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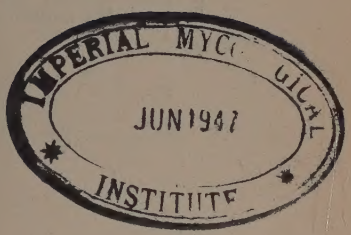

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MATHEMATICS

THE DUALITY LAW FOR THE GENERALIZED OPERATION OF TRANSLATION

By B. LEWITAN

(Communicated by A. N. Kolmogoroff, Member of the Academy, 27. V. 1944)

D. Raikoff has shown [1] that on commutative locally compact groups with the second enumerability axiom Pontrjagin's duality law follows from Plancherel's theorem.

In the present note I will show that for every operation of generalized translation there may be constructed an operator of generalized translation dual to it [4–6], and that for such dual operations the duality law holds under certain additional conditions (which on groups are always satisfied).

1. Let T^s be a family of operators of generalized translation on Z (cf. [4]). Then, as has been shown before [6], there exists on the set of maximal ideals of the ring R such a totally additive measure $F(\Delta)$ that for arbitrary $E_1(\alpha)$, $E_2(\alpha) \in L_2$ holds the equality

$$\int E_1(\alpha)\overline{E_2(\alpha)}\,dF(\alpha) = \int f_1(t)\overline{f_2(t)}\,dm(t) \tag{1}$$

where

$$f_i(t) = \int E_i(\alpha)\,\varphi(\alpha, t)\,dF(\alpha) \qquad (i = 1, 2)$$

For an arbitrary function $E(\alpha) \in L_2$ put

$$\tilde{S}_\alpha^\beta E(\alpha) = \int \overline{\varphi(\alpha, t)}\,\overline{\varphi(\beta, t)}\,f(t)\,dm(t),$$

$$f(t) = \int E(\alpha)\,\varphi(\alpha, t)\,dF(\alpha)$$

(convergence by the norm). From the equation (1) follows that the conditions of generalized translation II, III, IV and partially V are satisfied for the so defined operator of generalized translation. Whether the remaining conditions of generalized translation are satisfied or not, is probably connected with the deeper properties of the function $\varphi(\alpha, t)$. Here we will confine ourselve- to the formal side of the matter and assume correspondingly that all cons ditions of generalized translation are satisfied.

For the operation of generalized translation S^β we construct the ring R' with the multiplication

$$E * G(\alpha) = \int \widetilde{S}_\alpha^\beta E(\alpha)\, G(\beta)\, dF(\beta) =$$
$$= \int G(\beta) \left[\int \overline{\varphi(\alpha, t)}\, \varphi(\beta, t)\, f(t)\, dm(t) \right] dF(\beta).$$

Let us show that every function $\varphi(\alpha, t)$ generates a maximal ideal in the ring R'. To this end it suffices to put

$$(E, M) = \int E(\alpha)\, \varphi(\alpha, t)\, dF(\alpha)$$

since in virtue of the inversion formulae

$$(E * G(\alpha), M) = \int G(\beta)\, dF(\beta) \int\int \overline{\varphi(\alpha, t)}\, \varphi(\beta, t)\, f(t)\, dm(t)\, \varphi(\alpha, z)\, dF(\alpha) =$$
$$= \int G(\beta)\, f(z)\, \varphi(\beta, z)\, dF(\beta) = \int G(\beta)\, \varphi(\beta, z)\, dF(\beta) \int E(\beta)\, \varphi(\beta, z)\, dF(\beta) =$$
$$= (E, M)(G, M)$$

Let the set of all maximal ideals of the ring R' fill a certain space Z' so that $Z \subset Z'$. Let $m'(\Delta')$ be the measure generated by the operators S^β on Z'. By Plancherel's theorem

$$\int_Z |f(t')|^2\, dm'(t') = \int |E(\alpha)|^2\, dF(\alpha) \tag{2}$$

for every $f(t') \in L_2$.

Theorem. *If $m'(Z' - Z) > 0$ and if the ring R' is regular (i. e. for every point t' and its neighbourhood V' there exists a function $E(\alpha) \in L_1$ such that*

$$\int E(\alpha)\, \varphi(\alpha, t')\, dF(\alpha) \neq 0 \quad \text{and} \quad \int E(\alpha)\, \varphi(\alpha, t')\, dF(\alpha) = 0$$

outside V'), then $Z'' = Z$, i. e. the law of duality holds.

Proof. Let $t' \in Z' - Z$. Since $Z' - Z$ is a difference of closed sets, it is an open set. Consequently, t' enters into $Z' - Z$ together with its neighbourhood V'. Let $m(V') > 0$ (for at least one neighbourhood V' this will be true). By assumption there exists such a function $E(\alpha)$ that

$$\int E(\alpha)\, \varphi(\alpha, t')\, dF(\alpha) = 0$$

outside V'. Consequently, the equality is all the more valid for all $t' \in Z$.

By Plancherel's theorem (for the ring R)

$$\int |E(\alpha)|^2\, dF(\alpha) = 0$$

Hence

$$\int |E(\alpha)|\, dF(\alpha) = 0, \quad \int E(\alpha)\, \varphi(\alpha, t')\, dF(\alpha) = 0$$

which contradicts the condition $\int E(\alpha)\, \varphi(\alpha, t')\, dF(\alpha) \neq 0$ for $t' \in V'$.

It was shown in Raikoff's note (1) that in the case of groups the conditions of the theorem are satisfied.

2. Here is an example of dual rings, which we state without proof. Consider the differential operator $d^2u/dt^2+\rho(t)u=L_t(u)$, $\int_0^\infty |\rho(t)|\,dt<\infty$.

Define the operation of generalized translation $T_t^s f(t)=\Phi(s,t)$ as the solution of the equation $L_s\Phi=L_t\Phi$ with the initial conditions

$$\Phi(0,t)=f(t),\ \partial/\partial t\cdot\Phi(s,t)|_{s=0}=0.$$

By means of Riemann's function this operation may be extended to arbitrary measurable functions.

Povsner (2) stated a series of conditions for T^s to be an operation of generalized translation. In this case the maximal ideals will be generated by the solutions $\varphi(\lambda,t)$ of the equation $L(\varphi)=-\lambda^2\varphi$. Applying the results of the foregoing note, we obtain a decomposition of arbitrary functions into integrals of Fourier type

$$f(s)=\int_0^\infty e(\lambda)\varphi(\lambda,s)\,d\lambda,\quad e(\lambda)=\int_0^\infty f(s)\varphi(\lambda,s)\,ds \tag{3}$$

where $f(s)$, $e(\lambda)\in L_2$, the convergence being taken by the norm.

The first of these formulae has been already proved in a more general case by Weyl (3). As to the second formula, it is, as I have shown, a corollary of Green's formula. Define now the operator of generalized translation S^μ by setting

$$S_\lambda^\mu e(\lambda)=\int_0^\infty f(s)\varphi(\lambda,s)\varphi(\mu,s)\,ds$$

where $f(s)$ is determined by formula (3).

The proof that the operators satisfy all conditions of generalized translation may be carried out in this case by using the asymptotic formulae for $\varphi(\lambda,s)$. Observe that the operators S^μ may be extended to the aggregate of all measurable bounded functions by the summation method. This extension as well as the study of almost periodic functions for the operators S^μ is carried out elsewhere (7).

The F. Dzerjinski Artillery Academy.

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MATHEMATICS

CONSTRUCTION OF FIELDS OF ALGEBRAICAL NUMBERS WHOSE GALOIS GROUP IS A GROUP OF QUATERNION UNITS

By D. K. FADDEYEV

(Communicated by I. M. Vinogradow, Member of the Academy, 27. VII. 1944)

1. This paper has for its object the construction of fields of algebraical numbers whose Galois group over a given field $\mathbf{R}$ is a group of order 24 isomorphic to the (multiplicative) group formed by the quaternions ± 1, $\pm i$, $\pm j$, $\pm k$, $\frac{1}{2}(\pm 1 \pm i \pm j \pm k)$. This group has a unique principal series $Q_{24} \supset Q_8 \supset Q_2 \supset E$. Here Q_8 is a group formed by the elements ± 1, $\pm i$, $\pm j$, $\pm k$; Q_2 is formed by ± 1; Q_2 is the centre of Q_{24}; $Q_{24}/Q_8 \approx \mathfrak{A}_3$ is a cyclic group of order 3; $Q_{24}/Q_2 \approx \mathfrak{A}_4$ is the tetrahedron group. The following semi-direct decomposition takes place: $Q_{24} = Q_8 \cdot F$; F is a cyclic (not invariant) subgroup of order 3, the elements of which are 1, ε, ε^2, where $\varepsilon = \frac{1}{2}(-1+1+j+k)$. We have $\varepsilon^{-1} i \varepsilon = j$, $\varepsilon^{-1} j \varepsilon = k$, $\varepsilon^{-1} k \varepsilon = i$.

Let $\mathbf{K}$ be a normal field whose Galois group over the given field $\mathbf{R}$ is the group Q_{24}. Denote by $\mathbf{k}$ and $\mathbf{k}_1$ the subfields of $\mathbf{K}$ belonging to the subgroups Q_8 and Q_2, respectively; $\mathbf{k}$ is a cubic cyclic field, $\mathbf{k}_1$ is the field whose Galois group is the tetrahedron group. Consider the quaternion $r = \sum_{\sigma \in Q_{24}} x^\sigma \sigma$, where x is an element of $\mathbf{K}$, which forms together with the conjugate elements x^σ a basis of $\mathbf{K}$; σ denotes an automorphism of the field $\mathbf{K}$, as well as the corresponding quaternion.

It is evident that, for every $\tau \in Q_{24}$,

$$r^\tau = r\tau^{-1} \qquad (*)$$

Consider now the quaternion $rir^{-1} = \xi_1 i + \xi_2 j + \xi_3 k$. In virtue of (*) this quaternion is invariant under the substitutions ± 1, $\pm i$, and it changes the sign under the substitutions $\pm j$, $\pm k$. Let ξ be a non-zero linear combination of the numbers ξ_1, ξ_2, ξ_3. Multiplying and dividing by ξ we obtain $rir^{-1} = \frac{1}{\xi}(\alpha_1 i + \alpha_2 j + \alpha_3 k)$, where α_1, α_2, α_3 rest invariant under all the substitutions of the group Q_8 and, consequently, belong to the field $\mathbf{k}$.

Thereby $\lambda = \xi^2 \in \mathbf{K}$, but $\xi \in \mathbf{K}$. Since the norm of i is 1, $\lambda = \alpha_1^2 + \alpha_2^2 + \alpha_3^2$. The substitution transforms the quaternion rir^{-1} into $r\varepsilon^{-1} i \varepsilon r^{-1} = rjr^{-1}$. On the other hand, the substitution ε transforms the numbers of the field $\mathbf{k}$ into conjugate numbers. Consequently, $rjr^{-1} = \frac{1}{\sqrt{\lambda}}(\alpha_1' i + \alpha_2' j + \alpha_3' k)$. Since the quaternions i and j are orthogonal, rir^{-1} and rjr^{-1} are orthogonal, too. Consequently, $\alpha_1\alpha_1' + \alpha_2\alpha_2' + \alpha_3\alpha_3' = 0$.

Now let us show that, given any three elements α_1, α_2, α_3 of cubic cyclic field $\mathbf{k}$ that satisfy the conditions $\alpha_1\alpha_1' + \alpha_2\alpha_2' + \alpha_3\alpha_3' = 0$, $\sqrt{\alpha_1^2 + \alpha_2^2 + \alpha_3^2} \in \mathbf{k}$, we can construct a field with the group Q_{24}. We take the quaternions $\alpha_1 i + \alpha_2 j + \alpha_3 k$, $\alpha_1' i + \alpha_2' j + \alpha_3' k$ and normalize them. Notice that the coordinates of the quaternions so obtained belong to a tetrahedric field $\mathbf{k}_1$. For, $N(\lambda) = N(\alpha_1^2 + \alpha_2^2 + \alpha_3^2) = k^2$, where k is the determinant consisting of α_1, α_2, α_3 and the conjugate numbers, which is, evidently, a number from the fundamental field $\mathbf{R}$. Consequently (see [2] chapt. III), the Galois group of the field $\mathbf{k}_1 = \mathbf{k}(\sqrt{\lambda}, \sqrt{\lambda'})$ is the tetrahedron group $\mathfrak{A}_4$. Denote by i', j', k' the substitutions of this group, which leave invariant $\sqrt{\lambda}$, $\sqrt{\lambda'}$, $\sqrt{\lambda''}$, respectively. By ε' we denote the substitution which transforms $\sqrt{\lambda}$ and $\sqrt{\lambda'}$ into $\sqrt{\lambda'}$ and $\sqrt{\lambda''}$.

In this way we establish an isomorphic correspondence between the substitutions of the Galois group of the field $\mathbf{k}_1$ and the elements of the quotient-group Q_{24}/Q_2. We choose the quaternion r' so as to have $r' i r'^{-1} = \frac{1}{\sqrt{\lambda}}(\alpha_1 i + \alpha_2 j + \alpha_3 k)$, $r' j r'^{-1} = \frac{1}{\sqrt{\lambda'}}(\alpha_1' i + \alpha_2' j + \alpha_3' k)$. This is always possible, the right-hand sides being orthonormalized. Since the problem is reduced to a system of linear equations, the coefficients of r' can be chosen in $\mathbf{k}_1$. The quaternion r' is determined uniquely, up to a scalar factor. It is easy to see that, under every substitution σ' of the Galois group of $\mathbf{k}_1$, the quaternions $r' i r'^{-1}$ and $r' j r'^{-1}$ vary in the same manner as on multiplication of r' by the corresponding quaternion σ.

Consequently, $r'^{\sigma} \sigma r'^{-1}$ is a scalar, namely, a number from $\mathbf{k}_1$.

Introduce the notation $r'^{\sigma} \sigma r'^{-1} = \rho_\sigma$. It is evident that

$$\rho_{\sigma_1}^{\sigma_2} \rho_{\sigma_2} = \rho_{\sigma_1 \sigma_2}, \quad \rho_{-1} = -1 \qquad (**)$$

Consider a number $\mu = a|r'|^2$, where a is an arbitrary number from $\mathbf{R}$. It is easy to see that the Galois group of the field $\mathbf{K} = \mathbf{k}_1(\sqrt{\mu})$ is the group Q_{24}. For the field $\mathbf{k}_1(\sqrt{\mu})$ is normal, because $\mu^{\sigma'} = \mu \rho_\sigma^2$, and its Galois group is formed by the substitutions carrying $\sqrt{\mu}$ into $\pm\sqrt{\mu^{\sigma'}}$, $\sigma' \in \mathfrak{A}_4$. Denote by S_σ the substitution that transforms $\sqrt{\mu}$ into $\rho_\sigma \sqrt{\mu}$. Then, in virtue of (**), $S_{\sigma_1} S_{\sigma_2} = S_{\sigma_1 \sigma_2}$, *i. e.* the Galois group of the field $\mathbf{K}$ is isomorphic to the group Q_{24}.

2. Thus, the problem to construct the fields with the group Q_{24} is reduced to the determination of triplets of numbers α_1, α_2, α_3 in a cubic cyclic field $\mathbf{k}$ that possess the properties $\alpha_1\alpha_1' + \alpha_2\alpha_2' + \alpha_3\alpha_3' = 0$, $\sqrt{\alpha_1^2 + \alpha_2^2 + \alpha_3^2} \in \mathbf{k}$. Let us prove the existence of such triplets in every cubic cyclic field $\mathbf{k}$. Suppose $u = \alpha_1 i + \alpha_2 j + \alpha_3 k$ be a quaternion, the coordinates of which satisfy the first condition, and μ is an arbitrary fixed number in $\mathbf{k}$ which does not belong to the fundamental field. Let further $t = \mu + \mu'\varepsilon + \mu''\varepsilon^2$, where $\varepsilon = \frac{1}{2}(-1 + i + j + k)$. Take the quaternion $v = t^{-1} u t = \beta_1 i + \beta_2 j + \beta_3 k$. Then $v' = t'^{-1} u' - t' = \varepsilon t^{-1} u' t \varepsilon^{-1}$, and, u and u' being orthogonal, $\varepsilon^{-1} v' \varepsilon = \beta_1' j + \beta_2' k + \beta_3' i = t^{-1} u' t$ is orthogonal to $v = t^{-1} u t$.

Conversely, having a quaternion v orthogonal to $\varepsilon^{-1} v' \varepsilon$, we obtain, by putting $u = tvt^{-1}$, a quaternion u orthogonal to u', as is required. As to the quaternion v, it can be found by solving an intermediate equation $\beta_1\beta_3' + \beta_2\beta_2' + \beta_3\beta_2' = 0$ in the numbers of the field $\mathbf{k}$. This equation can be reduced to the system of linear equations $\beta_2'\beta_3 + \beta_1\beta_3' = -\beta_2\beta_1'$, $\beta_2''\beta_3' + \beta_1'\beta_3'' = -\beta_2'\beta_1''$, $\beta_1''\beta_3 + \beta_2\beta_3'' = -\beta_2''\beta_1$ with respect to β_3, $\beta_3'\beta_3''$. Thereby we may assign to β_1 and β_2 any values from $\mathbf{k}$ provided the determinant of the system $\beta_1\beta_1'\beta_1'' + \beta_2\beta_2'\beta_2''$ be different from zero. The numbers β_3, β_3', β_3'' assume conjugate values from $\mathbf{k}$. Since for at least one pair out of the numbers β_1, β_2, β_3 the

sum of the norms is different from zero, no triplet will be omitted when this method of construction is applied.

It is not difficult to compute that for β_1 and β_2 indeterminate, $\alpha_1^2+\alpha_2^2+\alpha_3^2=\beta_1^2+\beta_2^2+\beta_3^2$ is not a square. Hence, by Hilbert's theorem, there are infinitely many values of β_1, β_2 for which $\alpha_1^2+\alpha_2^2+\alpha_3^2$ is not the square of any number from $\mathbf{k}$. Thus, an arbitrary cubic cyclic field $\mathbf{k}$ can be imbedded in a field with the group Q_{24}.

3. Now we shall give an arithmetical criterion for a tetrahedric field $\mathbf{k}_1$ to be imbeddable in a field with the group Q_{24} in case the fundamental field $\mathbf{R}$ is the field of rational numbers.

Let $\mathbf{k}_1=\mathbf{k}(\sqrt{\lambda}, \sqrt{\lambda'})$ be a field with the tetrahedron group. Here λ is a number from a cubic cyclic field $\mathbf{k}$, the norm of which is a square. Without restraining the generality we may suppose λ to be integral. Consider the algebra A formed by the quaternions $c+\alpha\sqrt{\lambda}i+\alpha'\sqrt{\lambda'}j+\alpha''\sqrt{\lambda''}k$, where c runs over the rational numbers, α runs over the numbers of the field $\mathbf{k}$. It is easily seen that A is simple and normal over $\mathbf{R}$, *i. e.* A is the algebra of generalized quaternions. The field $\mathbf{k}_1$ is imbeddable in a field with the group Q_{24} if and only if $\lambda=\alpha_1^2+\alpha_2^2+\alpha_3^2$, where $\alpha_1\alpha_1'+\alpha_2\alpha_2'+\alpha_3\alpha_3'=0$. In case this condition is fulfilled, the quaternions $r_s=(\alpha_s/\sqrt{\lambda})\,i+(\alpha_s'/\sqrt{\lambda'})\,j+(\alpha_s''/\sqrt{\lambda''})\,k$, $s=1, 2, 3$, contained in A are orthogonal and normalized, *t. e.* A is equivalent to the ordinary quaternion algebra. Conversely, if A is equivalent to the ordinary quaternion algebra, then, as it is easy to see, λ admits the required representation.

By Hasse-Brauer-Noether's theorem, the algebra A is equivalent to the ordinary quaternion algebra if and only if A decomposes over all the odd primes and does not decompose over ρ_∞. The last condition is equivalent to the positiveness of λ, together with the conjugate numbers.

Suppose $[\lambda]=\gamma^{\sigma_3}$, $N(\gamma)=a^2$, and the different ϑ of the field $\mathbf{k}$ is f^2. Then, as is easily seen, the elements $c+\alpha\sqrt{\lambda}i+\alpha'\sqrt{\lambda''}j+\alpha''\sqrt{\lambda'}k$ with c running over the integral rational numbers, while α runs through the ideal $1/f\delta$, form the order of the algebra, the discriminant of which is equal to $-16a^2$. Consequently, only the divisors of the number $2a$ can be critical prime numbers of the algebra.

The number λ can be chosen in such a way that the ideal δ is prime both to γ and to the discriminant of the field $\mathbf{k}$. Let λ be a root of the equation $\lambda^3-q\lambda^2+t\lambda-a^2b^2=0$. It is easy to see that the coefficient t is divisible by a, $t=as$ and $(s, a)=(q, a)=1$.

Consider the quaternions $\rho_1=\sqrt{\lambda}i+\sqrt{\lambda'}j+\sqrt{\lambda''}k$ and $\rho_2=(\lambda'-\lambda'')\sqrt{\lambda}i+(\lambda''-\lambda)\sqrt{\lambda'}j+(\lambda-\lambda')\sqrt{\lambda''}k$. It is easily seen that $\rho_1^2=-q$, $\rho_2^2=-Q=-a(sq-9ab^2)$, $\rho_2\rho_1=-\rho_1\rho_2$, *i. e.* the algebra A is the cross-product of the field $\mathbf{R}(\sqrt{-Q})$ by its group with the factor ρ_1 whose square is $-q$. This algebra decomposes over all the odd prime divisors ρ_a of the number a, *i. e.* A is equivalent to the ordinary quaternion algebra if and only if $(-q/p_a)=+1$.

Thus, the following conditions are necessary and sufficient that the tetrahedric field $\mathbf{k}(\sqrt{\lambda}, \sqrt{\lambda'})$ could be imbedded in a field with the group Q_{24}: 1) λ and all its conjugate numbers are positive; 2) if $\lambda=\gamma\delta^2$, $N(\gamma)=a^2$, $(ad, \delta)=1$, $\lambda+\lambda'+\lambda''=q$, then $(-q/p_a)=1$ for all odd prime divisors of a.

The Leningrad Department of the Stekloff Mathematical Institute. Academy of Sciences of the USSR.

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MECHANICS

DIAGRAM OF ANGULAR VELOCITIES OF A REGULAR TOOTH GEAR MECHANISM WITH TWO DEGREES OF FREEDOM

By M. KREINES and M. ROZOVSKY

(Communicated by L. S. Leibenson, Member of the Academy, 28. I. 1945)

Let us adopt the following definitions:

1) Any mechanism consisting of several pairs of circular gears in mesh having one common planet carrier revolving about an axis immovable in space is said to be a differential tooth-gear mechanism.

2) Any link of this mechanism having its axis of rotation fixed in space may be treated as a main link.

3) A tooth-gear mechanism is said to be regular (1) if it is composed * of several differential mechanisms, in which some of the links can be made immovable.

It may be remarked that among the tooth-gear mechanisms used in modern machine-building industry a vast majority are regular.

The present note is devoted to the construction and investigation of the diagram of angular velocities for a regular tooth-gear mechanism with two degrees of freedom.

Let 0, n, 1, 2, ..., $n-1$ denote the driving, the fixed and the remaining links of the mechanism under consideration. Seeing that our mechanism has two degrees of freedom and that, being regular, it has the angular velocities ω_0, ω_1, $\omega_2, \ldots, \omega_n$ of its main links 0, 1, 2, ..., n bound by linear and homogeneous relations with constant coefficients, we can write down these relations in the form:

$$\begin{aligned}
\omega_0 &= a_0 u + b_0 v \\
\omega_1 &= a_1 u + b_1 v \\
\omega_2 &= a_2 u + b_2 v \\
&\cdots\cdots\cdots \\
&\cdots\cdots\cdots \\
\omega_{n-1} &= a_{n-1} u + b_{n-1} v \\
\omega_n &= 0
\end{aligned} \qquad (1)$$

where u and v are two independent parameters; $a_0, a_1, a_2, \ldots, a_{n-1}$ and $b_0, b_1, b_2, \ldots, b_{n-1}$ are constant coefficients depending upon the construction of the mechanism and the choice of the parameters u and v.

Note that among the $\omega_0, \omega_1, \omega_2, \ldots, \omega_{n-1}$ any two linearly independent angular velocities may be chosen for u and v. In the sequel we put $v = \omega_0$, and in measuring angular velocities put the angular velocity of the driving gear at unity. The system (1) will then take the form

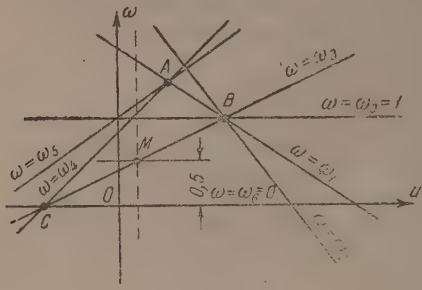
* A mechanism A is said to be composed of mechanisms B and C, if A is formed by establishing a rigid connexion between some main coaxial links of B and C.

$$\begin{aligned}
\omega_1 &= a_1 u + b_1 \\
\omega_2 &= a_2 u + b_2 \\
&\cdots\cdots\cdots \\
&\cdots\cdots\cdots \\
\omega_{n-1} &= a_{n-1} u + b_{n-1} \\
\omega_n &= 0
\end{aligned} \qquad (2)$$

By means of equations (2), graphs of angular velocity for all the main links of the mechanism may be plotted in the rectangular system of coordinates $uO\omega$. Each of these plots ($\omega = \omega_0$, $\omega = \omega_1$, $\omega = \omega_2$, . . . , $\omega = \omega_n$) will be evidently a straight line.

The whole may be described as the angular velocity diagram of the mechanism under consideration.

In the adjoined figure we have this diagram plotted for a regular mechanism composed of 7 main links.



As may be easily seen, the diagram of angular velocities enjoys the following properties:

I. Given the angular velocity of one main link, we can by means of this diagram find the angular velocities of all the other main links. Taking the diagram of the figure as an example, suppose we are given the velocity ω_3 equal, say, to 0.5 ω_0. Then in order to determine the angular velocities of the other links, we find on the line $\omega = \omega_3$ the point M with the ordinate $\omega = 0.5$. The ordinates of those points of the remaining straight lines which are situated on one vertical with the point M will give us the respective values of angular velocities for the other links.

II. The relative angular velocities of any two main links p and q with respect to each other will be equal to the difference between the ordinates of those points of the lines $\omega = \omega_p$ and $\omega = \omega_q$ which have the same abscissa u, corresponding to the considered regime of the mechanism work.

III. The diagram shows that in a regular mechanism with two degrees of freedom the angular velocities of its different main links attain their extremal values simultaneously (these extremal values correspond to the extremal values of the parameter u).

IV. Suppose that beside the driving link only two arbitrary links p and q of the mechanism have moments applied to them externally, and that the angular velocities ω_0, ω_p and ω_q are pairwise linearly independent.

Then, using the diagram, we may easily determine the moments M_p and M_q applied to the links p and q.

Indeed, it may be shown that

$$M_p = -\frac{1}{\omega_p|_{\omega_q=0}} M_0 \text{ and } M_q = -\frac{1}{\omega_q|_{\omega_p=0}} M_0$$

where M_0 is the exterior moment applied to the driving link 0.

V. For any differential mechanism with two degrees of freedom, which is a part of the considered regular mechanism, the graphs of angular velocities of all its main links intersect on the diagram at one point (cf. the points A, B and C* on the figure).

---

* In the differential mechanism with two degrees of freedom corresponding to the point C, one of the links—the link 6—has been made immovable. Accordingly, this mechanism has become a system with one degree of freedom.

The gear ratio between one main link of such a regular mechanism and another main link, when the planet carrier is stopped, is equal to the ratio of two directed segments of an arbitrary vertical line. Each of these segments has its initial point at the intersection of the vertical with the graph of angular velocity of the planet carrier of the differential mechanism, while its end-point lies at the intersection of this vertical with the graphs of angular velocities of the two remaining links.

Every regular mechanism with two degrees of freedom has its angular velocity diagram consisting of several straight lines. There naturally arises the inverse question: is it possible to assign to every system of straight lines in the $uO\omega$-plane a regular mechanism with two degrees of freedom, for which this system of lines is its angular velocity diagram?

To answer this question consider a certain system of straight lines $\omega=\omega_0=1$, $\omega=\omega_n=0$, $\omega=\omega_1$, $\omega=\omega_2$, . . . , $\omega=\omega_{n-1}$, and note that between three quantities ω_p, ω_q and ω_r the relation $\omega_p-\omega_r=i_k(\omega_q-\omega_r)$, where i_k is a constant, holds if and only if the three $\omega=\omega_p$, $\omega=\omega_q$ and $\omega=\omega_r$ intersect at one point. Those points in our figure at which three or more given straight lines intersect will be described as knot-points, and we are to consider the system S of equations of the form $\omega_p-\omega_r=i\ (\omega_q-\omega_r)$ generated by different knot-points; it is easily seen that *for the considered system of straight lines a regular gear mechanism with two degrees of freedom, having this system for its angular velocity diagram, can be constructed then and only then when the system of equations S contains $n-2$ independent equations**.

The construction of such a mechanism is carried out on the basis of the following considerations.

It may be easily shown that from any number of main links having their angular velocity graphs intersecting at one point, a differential mechanism can be constructed that establishes between the angular velocities of these links the relations generated by the knot-point under consideration.

Any regular gear mechanism consisting of a certain number of these differential mechanisms and generating $n-2$ independent relations will be found to establish all necessary connexions between the $n+1$ links under consideration.

A fairly large number of such mechanisms, differing in construction, may be composed. Every one of them can be derived from the diagram of angular velocities.

Thus, the synthesis of regular gear mechanisms is considerably simplified by the use of angular velocity diagrams.

In constructing a regular gear mechanism with two degrees of freedom we must, according to the purpose which this mechanism is intended to serve, determine the number of necessary main links and establish the equations between their angular velocities. If after drawing the angular velocity graphs for these links it turns out that the totality of the obtained straight lines is no diagram of angular velocities of any regular gear mechanism with two degrees of freedom, then it is necessary, in order to make the obtained diagram (we shall call it the basic diagram) an angular velocity diagram of some mechanism, to introduce additional straight lines taking them for angular velocity graphs of additional links of the mechanism to be constructed.

By means of the angular velocity diagram it is possible in certain cases easily to determine the minimum number of necessary additional links, to choose them moreover in the most judicious manner, since in many cases not merely the velocities but also the moments can be derived from the angular velocity diagram.

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* The i_k are, of course, assumed to be rational numbers.

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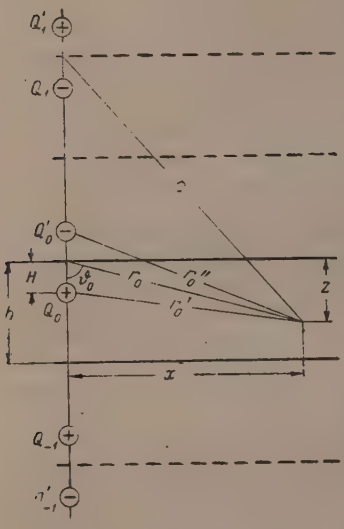
PHYSICS

SOUND RADIATION FROM A SOURCE PLACED IN WATER AT A SMALL DEPTH

By L. M. BREKHOVSKIKH

(Communicated by S. I. Wawilow, Member of the Academy, 11. X. 1944)

A sound radiator placed in water at a small depth has always its power more or less weakened by its proximity to the surface. There arises the essential problem of how deep should the source be submerged in order that this weakening might be neglected. In the present paper we give the solution of this problem and of some other related to it.



1. Deep Sea. In the first place the calculation is made for the case where the depth of the sea is large in comparison with the distance from source to receiver, so that the influence of reflections from the bottom might be disregarded.

The same state of affairs will take place if the sea is not deep, but the bottom reflects the sound feebly. In such cases only the direct ray and the ray reflected from the surface of the sea reach the receiver. The reflected ray can be supposed to issue from an imaginary source which is the mirror image of the real source reflected on the surface of water (which we consider to be a plane surface). Seeing that reflection from the water surface is attended by a jump in the wave-phase equal to π, the imaginary source will work 180° out of phase (in counterphase) with the real one.

Denote by H the depth of immersion of the source; by z, the depth of immersion of the receiver; by r_0, the distance of the receiver from that point of the water surface which lies above the source; by ϑ_0, the angle between the direction of r_0, and the water surface (see figure).

Suppose that $H \ll r_0$, $z \ll r_0$. On this assumption the two sources can be considered to lie at nearly equal distances from the receiver, and therefore, the sound pressure created by them will be

$$p_0 = \frac{A}{r_0}\left[\cos\left(\omega t - kr_0 - \frac{1}{2}kl\right) - \cos\left(\omega t - kr_0 + \frac{1}{2}kl\right)\right] \qquad (1)$$

where A is expressed in terms of the volume velocity of the source, which we always suppose to be unity; l is the path difference between the sources

$$l = 2H \sin \vartheta_0 = \frac{2Hz}{r_0} \tag{2}$$

$k = \omega/c = 2\pi/\lambda$ is the wave number.

For the square mean value of pressure we get from (1)

$$p = \frac{\sqrt{2}A}{r_0} \sin \frac{kl}{2} \cong \frac{\sqrt{2}AHkz}{r_0^2} \tag{3}$$

It will be obvious that the pressure varies directly as the depth of the source, and is also proportional to the depth of the receiver. It may be seen from (3) that this conclusion holds as long as $kzH/r_0 \ll 1$. The same expression shows that the increase stops with $kl = \pi$, or, taking only account of the order of magnitude, when $kzH/r_0 \cong \pi/2$. If the depth of the receiver is approximately equal to $\frac{1}{4}$ of the wave-length, *i. e*, $kz \sim \pi/2$, it will be apparent that the increase in pressure stops at a depth H approximately equal to the distance r_0 from source to receiver.

The above results refer to the case where the pressure created by the source is considered at points lying in directions nearly grazing the surface of the water ($z/r_0 \ll 1$). Things will be different if we are to consider the integral power radiated by the source. For this purpose let us take the square of the expression (3); expand $\sin^2 \frac{kl}{2}$ in a power series with respect to the argument; substitute $l = 2H \sin \vartheta_0$ and integrate over a half of the solid angle. Comparing the integral power W thus obtained with the power W_0 radiated by the source in an infinite space of water, we obtain the formula

$$W = \frac{2W_0}{3}\left(k^2H^2 - \frac{k^4H^4}{5}\right) \tag{4}$$

Neglect of $(kH)^6$ and of the following term will give an error not exceeding 10 per cent if $kH \leqslant 1$. If $kH = 1$ (which corresponds to $H = \lambda/2\pi$) we have $W \cong W_0/2$. It will be apparent that in order for as much as half of the power to be emitted it will suffice to submerge the source to a depth equal to 1/6 of the wave-length.

To end with this case, let us note an important conclusion arising from (3). The sound pressure decreases with the distance (for z constant) as $1/r_0^2$.

2. Finite Depth and Absolutely Reflecting Bottom. This is the extreme opposite of the case which we have just discussed. In our computation let us make use of the fact that the successive reflections of the sound from the bottom and surface can be replaced by a sound coming in from a chain of imaginary sources, and as the reflections from the bottom take place without any change in the phase of the wave, while the reflections from the water surface are attended by a jump of the phase as large as π, the sources in the chain will be arranged in pairs with opposite phases. In the figure Q_0 is the real source; Q_0', Q_1, Q_1', ... are imaginary sources. Denote by h the depth of the sea; by x, the distance from source to receiver along the horizontal. Suppose that $H \ll h$, $h \ll x$. The first condition means that the source is near the surface of the water. Owing to this, all sources in the chain are arranged pairwise. Denote by r_m the distance between the pair Q_m, Q_m' and the receiver. It is easy to show that if the bottom is absolutely reflecting, the sound field in water can be substituted with a field generated by such a chain of sources. In other words, both the differential equation of the field and the boundary conditions at the bottom and at the water surface are satisfied.

It is not possible, however, to compute the total field of all sources accurately; mainly, because of the different delay in radiation coming in from different sources. In carrying out the computation we will make use of the fact that one has more often to deal with non-monochromatic sources of sound, *i. e.* with such as have a finite coherence length, so that if the path difference between the sources is larger than the above length, their energies can be summed up without taking into account the phases. If we suppose that in a radiation of equal intensity all frequencies from ω_1 to $\omega_1+\Delta$ are represented, then the coherence length will be

$$\tau = 2\pi/\Delta \tag{5}$$

In our computation we will suppose $c\tau$ to be smaller than the path difference between the two closest pairs of sources, *i. e.* between Q_0, Q_0' and Q_1, Q_1'. This implies the fulfilment of the following inequality:

$$\frac{4h^2}{x} \geqslant c\tau \tag{6}$$

H being small, the radiation from every pair of sources has to be summed up with allowance for the phase difference between them. For the square of the amplitude of pressure created by the pair Q_m, Q_m' we obtain, after a rather simple calculation, with an accuracy up to $(kH)^4$ the following expression:

$$p_m^2 = 4A^2 \left[\frac{kH\,(2mh+z)}{r_m^2}\right]^2 \left[1+\frac{(kH)^2\,(2mh+z)^2}{3r_m^2}\right] \tag{7}$$

This expression must be summed up over all m from $-\infty$ to $+\infty$. However, this summation can with sufficient accuracy be replaced by integration, since we have $x \gg h$ (2).

As a result we obtain for the square of the amplitude of the total pressure

$$p^2 = \frac{\pi A^2}{hx}\left[(kH)^2 - \frac{(kH)^4}{4}\right] \tag{8}$$

This formula is as accurate as (4). Again the pressure is zero for $H=0$ and increases with depth. We will describe as power of reference the power W_0 radiated by a source placed deep enough under water, say, when its position is in the middle of the water layer. In this case the source creates in the layer a «normal» sound pressure of amplitude p_0. In dealing with this case all sources may be considered to be mutually incoherent, and the same procedure as used above will give

$$p_0^2 = \frac{\pi A^2}{hx} \tag{9}$$

From (8) and (9) we get

$$p^2 = sp_0^2, \text{ where } s = (kH)^2 - \frac{(kH)^4}{4} \tag{10}$$

A similar result is obtained for the power radiated by the source, *i. e.*

$$W(H) = sW_0 \tag{11}$$

For $kH=1$ (which corresponds to $H=\lambda/2\pi$) we have $s=3/4$. For $kH=1.4$ ($H\cong\lambda/4$) we have $s=1$. In other words, the power of reference and the pressure corresponding to it are attained when the depth of the source reaches a value ranging from $\lambda/6$ to $\lambda/4$.

Until now we have done no averaging over the frequency, and, therefore, this conclusion applies only to radiators having a spectrum of small breadth, *i. e.* when $\Delta \ll \omega_0$, where ω_0 is the average frequency of the spectrum. The averaging (12) over the frequency is quite an elementary operation if the spectral characteristic of the source is given.

Often the octave analysis of radiation is used, *i. e.* the readings of the receiver are averaged over the range of an octave. If within the range of the octave under consideration the source has a uniform spectrum, the averaging is reduced to integration of s from ω_1 to $2\omega_1$ (ω_1 is the lower limit of the octave) with subsequent division by ω_1. As a result we obtain

$$\bar{s} = (k_0 H)^2 - \frac{1}{3.6}(k_0 H)^4 \qquad (12)$$

where $k_0 = 3\omega_1/2c$ corresponds to the mean frequency of the octave. Formula (12) is nearly identical with (10).

It may be remarked that, according to (8) and (9), the sound pressure varies inversely to the square root of the distance from the source, which for an absolutely reflecting bottom follows also from the law of conservation of energy.

3. Source in a Screen. Importance attaches to the case where a point-source is placed in a screen, *e. g.* in the side of a ship. To take the simplest case, we can suppose the point-source to be located at the surface of a hemisphere dipped into water. The size of the hemisphere will be assumed to be small in comparison with the wave-length. Adopting the image method, in the case of a deep sea we obtain two sources working in counter-phase and located symmetrically on the surface of the sphere. For a sea of finite depth we obtain a chain of such pairs.

The expression for the radiation of a point-source placed on a sphere is to be found in the literature ([3]). Using it, it is easy to show that the given pair of sources is in our approximation a dipole of certain effective length not coinciding with the distance between the sources, as measured along a straight line. This will mean that a source on a hemisphere can be replaced by a similar source located under a free surface of water at an effective depth H_{eff}. For the latter, computation gives

$$H_{eff} = \frac{3a}{2}\sin\varphi \qquad (13)$$

where a is the radius of the sphere, φ is the angle between the line connecting the centre of the sphere with the source, and the surface of the water. For $\varphi = \pi/2$ (the source is located at the lowest point of the hemisphere) we have $H_{eff} = 3a/2$, which nearly exactly coincides with $\pi a/2$, the shortest distance between the source and the free surface of water, as measured along the surface of the sphere.

Another interesting case is that of a piston-radiator located at the water surface, and whose one side is supposed to radiate into water. It is easy to see that by the image method this case is reduced to that of an oscillating disk whose both sides radiate sound waves. The solution of this case in our approximation has been given by Lamb ([4]). It turns out that a piston-radiator of radius a is equivalent to a point-radiator of order zero, having the same volume velocity and located at a depth a.

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PHYSICS

RADIATION OF A SOUND IN WATER AS AFFECTED BY DEPTH OF SUBMERSION

By N. N. ANDREYEV, Corresponding Member of the Academy, L. M. BREKHOVSKIKH and L. D. ROSENBERG

It has frequently been observed that the power of a sound source in water depends upon its depth of submersion. Schematically this phenomenon may be explained by the fact that when the source is near the surface, its pulsations give rise to a bulging rather than to condensations and rarefactions in the bulk of water.

A theoretical treatment of the problem by one of us [1] revealed that when a source of the zero order is submerged to a depth greater than one quarter of a wave-length corresponding to the mean frequency in its spectrum, there already will practically be no weakening in its radiation.

From the above considerations it is seen, in particular, that when two sources, one of zero order and the other of order one, are submerged to an equal depth, the weakening action of the water surface on the latter will be smaller, because the streamlines in this source are closed.

Not merely the total radiated power, but also the space distribution of the sound pressure created by the source depends upon the depth of submersion.

This is especially pronounced in very deep sea, where reflection from the bottom is practically absent. Suppose the source and the receiver are both near the surface.

Then the direct ray and the ray reflected from the surface may compensate each other, as they are mutually in counterphase. Therefore, in directions close to the horizontal the sound radiation is small. Depths of the source at which this will not take place, considerably exceed those required for maximum output of power and may approach the magnitude of the distances between the source and the receiver.

In shallow sea with highly reflecting bottom, where for every position of the receiver there is an infinite sequence of reflected rays, the picture is different. In this case the dependence of the power radiated and square of the sound pressure upon the depth of the source is the same for any position of the receiver.

To check the theory, a series of experiments was carried out in the sea under natural conditions. The sound source was a steel plate struck intermittently by a hammer (17 blows per second), which formed one side of a tank. The plate did not measure more than one metre along its largest side. The sound pressure produced was measured with the aid of a set-up consisting of a hydrophone, a band-pass-filter (breadth of a band is equal to an octave) and a calibrated amplifier with an output measuring instrument.

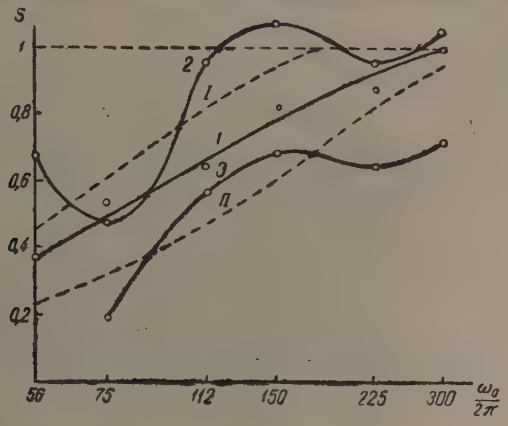
Absolute measurements of the sound pressure at short distances from the source when the latter was located at a sufficiently great depth, gave a practically uniform spectrum, approximately from 100 to 3000 cycles.

Two series of measurements were carried out, one at a sea depth of 8 m, and another at a sea depth of 6 m. The exact nature of the sea bottom at these points

was not known, but from certain data it could be concluded that it was rocky and covered with a thin layer of slime.

The method used was briefly the following. The source was set up at a definite depth, always no less than the draught of the ship, in order to minimize the influence of the hull on the sound field distribution. Sound pressure was measured in octaves at a distance of about 100 m with a hydrophone at a depth of 4 m. The pressure was measured in the following octaves: 37—75, 50—100, 75—150, 100—200, 150—300, 200—400, and so forth up to 12 000 cycles. Here we shall compare with theory only the results obained for the six octaves, since for higher frequencies the source already cannot be regarded as a point source. During measurements, the whole frequency range was traversed several times, and the mean value of all readings of the instrument for every octave was taken.

The first measurement was carried out at a source depth of 2 and 4 m. The results obtained are presented in the accompanying figure (curve *1*). The mean frequencies of the octaves have been plotted along the abscissae, using a logarith-



mic scale, and the ratio of the pressures measured at a source depth of 2 m to the pressures measured at a source depth of 4 m, *i. e.*, $s = p_{2m}/p_{4m}$, along the ordinates. We see that with an increase in frequency this ratio increases, approaching unity.

Exactly the same shape of the curve follows from the theoretical considerations. Indeed, for high frequencies a depth of 2 m is already quite sufficient to ensure a maximum output of power by the source, and increasing the depth to 4 m does not alter anything.

The theoretical dependence of the sound pressure upon the source depth for an absolutely reflecting bottom has the following form:

$$s = \frac{p}{p_0} = \sqrt{(k_0 H)^2 - \frac{(k_0 H)^4}{3.6}} \qquad (1)$$

where $k_0 = \omega_0/c = 2\pi/\lambda_0$ is the wave number corresponding to a mean frequency in the octave; H is the source depth; p is the sound pressure at the source depth H; p_0 is the pressure for the great depth of immersion (in our case, the pressure for the depth of 4 m).

Curve *I* in the figure shows the theoretical dependence of the ratio $s = p_{2m}/p_{4m}$ upon the frequency computed with the help of formula (1). We see that the expeperimental and theoretical curves practically coincide. Certain deviations here and there may be ascribed to experimental error.

In the second series of measurements the source was placed at a depth of 1, 2 and 3.5 m.

In the figure curve *2* shows the dependence of the ratio $p_{2m}/p_{3.5m}$, and curve *3*, the dependence of ratio $p_{1m}/p_{3.5m}$ upon the frequency. The first should

again be compared with the theoretical curve *I*, and the second, with curve *II* computed in a similar manner to curve *I*, but with $H = 1$ m.

On comparing the theoretical and experimental curves it is seen that the latter on the average have a tendency to follow the former, oscillating around them with a considerable amplitude. It is natural to explain these oscillations as due to the interference of sound waves after a diverse number of reflections from the bottom and surface, since, as can be easily checked, the condition for the absence of this interference (see(6)([1]))is not fulfilled here.

From a glance at the curves in the figure it is seen that the experimental curves, in a complete accord with the theoretical ones for an absolutely reflecting bottom, point to the fact that a maximum sound pressure is attained at a depth $H \cong \lambda_0/4$.

Consequently, an absolutely reflecting bottom is not an abstraction under actual conditions.

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GEOPHYSICS

A CONTRIBUTION TO THE THEORY OF TRANSFORMATION OF AIR-MASSES

By I. A. KIEBEL, Corresponding Member of the Academy

Although the problem of the transformation of an air-mass over a given surface belongs primarily in the domain of meteorology, its hydromechanical aspect is also of great interest. Considered from this angle, it is a problem of an unstationary boundary layer. There is, however, a circumstance which renders it more complicated in comparison with the problems of heat transfer as dealt with, for instance, in heat engineering. The state of affairs in the atmosphere of the Earth requires that the inflow of heat by radiation be considered alongside the inflow of heat by conduction (turbulent). Had the radiation been disregarded, it would have been impossible for us to discover the stratosphere by computation, just as the neglect of thermal conductivity would have prevented us from obtaining the distribution of temperature along the vertical, characteristic of the lower troposphere.

The inflow of heat in the atmosphere is very accurately described by the equation

$$c_p \rho \frac{dT}{dt} - \frac{c_p - c_v}{R} \frac{dp}{dt} = \varepsilon_1 + \varepsilon_2 \tag{1}$$

Here T is temperature; p, pressure; ρ, density; t, time; c_p and c_v, specific heat; R, gas constant; ε_1 and ε_2, the inflows of heat due to radiation and conduction, respectively*.

Assuming that only two flows of radiation are in existence: one directed downwards (including a long-wave radiation A and a short-wave, solar, radiation S) and another, directed upwards (long-wave radiation B), we can write, according to Schwarzshild and Emden,

$$\varepsilon_1 = \alpha \rho_w (A + B + \beta S - 2f\sigma T^4) \tag{2}$$

where α, $\beta\alpha$ and $f\alpha$ are absorption coefficients for grey radiation (α and $f\alpha$ for the long-wave radiation, $\beta\alpha$ for the short-wave radiation; the factor f is introduced according to Hulbert (1)); ρ_w, the density of the absorbing substance (water vapour, dust, *etc.*); σ, Stephan — Boltzmann's constant.

Confining ourselves in this paper to vertical mixing, we can write

$$\varepsilon_2 = \frac{\partial}{\partial z}\left(\lambda \frac{\partial T}{\partial z}\right) \tag{3}$$

where λ is the coefficient of turbulent thermal conductivity, z is the vertical coordinate. A, B and S satisfy the equations

$$\frac{\partial A}{\partial z} = \alpha \rho_w (A - f\delta T^4); \quad \frac{\partial B}{\partial z} = \alpha \rho_w (f\delta T^4 - B) \tag{4}$$

$$\partial s / \partial z = \beta \alpha \rho_w S \tag{5}$$

* The inflow of heat by evaporation is not taken into account.

The equations (1), (4), (5) in conjunction with the hydrodynamic equations of motion and continuity must solve the problem of transformation of air-mass. For boundary condition we take that with $z=0$ (at the surface of the Earth):

$$-\lambda\frac{\partial T}{\partial z}+\lambda^*\frac{\partial T}{\partial z}=A+S-B \tag{6}$$

$$B=q_f\sigma T^4 \tag{7}$$

Here λ^* and T^* are the coefficient of thermal conductivity and the temperature of the underlying surface (soil, water, *etc.*); q_f is a quantity whose value is close to unity *. For $z=\infty$ we can write

$$A=0 \tag{8}$$

$$B=W \quad \text{or} \quad \partial T/\partial t=0 \tag{9}$$

$$S=W \tag{10}$$

Here W is the flow of the Sun's radiation, multiplied by $1-\Gamma$; Γ is albedo.

Finally, for the new value T^* in (6) we have

$$c^*\rho^*\frac{\partial T^*}{\partial t}=\lambda^*\frac{\partial^2 T^*}{\partial z^2} \tag{11}$$

(where c^* and ρ^* are specific heat and density of the soil), the boundary conditions being

$$T^*=T \text{ for } z=0 \tag{12}$$

T^* is bounded for $z=-\infty$.

In an earlier paper ([2]) this system of equations has been solved for the stationary case where the left-hand side of (1) vanishes. In dealing with the problem of transformation, we leave on the left-hand side of (1) the term $(c_p\rho)\partial T/\partial t$.

Let us substitute the height z with the reduced optical thickness x as an independent variable:

$$x=\frac{1}{\tau_0}\int_z^\infty \alpha\rho_w dz \tag{13}$$

where $\tau_0=\int_0^\infty \alpha\rho_w dz$ **; instead of t introduce a dimensionsless time s: $s=\omega t$ ($1/\omega$ is the characteristic time). Further let us denote

$$A+B=a,\ B-A=y,\ f\varepsilon T^4=E \tag{14}$$

and introduce the quantities m and N from the equalities

$$\frac{4f\sigma\bar{T}^3}{\alpha\rho_w\lambda}=\frac{m^2-1}{2},\quad \frac{c_p\rho\omega}{4\alpha\rho_w f\sigma\bar{T}^3}=N \tag{15}$$

For a and y we can easily obtain the expressions:

$$a=-\frac{1}{\tau_0^2}\frac{\partial}{\partial x}\left(\frac{2}{m^2-1}\frac{\partial E}{\partial x}\right)+N\frac{\partial E}{\partial s}+2E-\beta S \tag{16}$$

$$y=-\frac{1}{\tau_0^3}\frac{\partial^2}{\partial x^2}\left(\frac{2}{m^2-1}\frac{\partial E}{\partial x}\right)+\frac{2}{\tau_0}\frac{\partial E}{\partial x}+\frac{1}{\tau_0}\frac{\partial}{\partial x}\left(N\frac{\partial E}{\partial s}\right)+\beta^2 S \tag{17}$$

* In order to simplify the problem, the flow of moisture in equation (6) is neglected.

** $\alpha\rho_w$ is taken to be independent of time.

Eliminating a and y (*i. e.*, A and B) from our system, we obtain the fundamental equation

$$\frac{1}{\tau_2^4}\frac{\partial^3}{\partial x^3}\left(\frac{2}{m^2-1}\frac{\partial E}{\partial x}\right)-\frac{1}{\tau_0^2}\frac{\partial}{\partial x}\left(\frac{2}{m^2-1}\frac{\partial E}{\partial x}\right)-\frac{2}{\tau_0^2}\frac{\partial^2 E}{\partial x^2}-$$
$$-\frac{1}{\tau_0^2}\frac{\partial^2}{\partial x^2}\left(N\frac{\partial E}{\partial s}\right)+N\frac{\partial E}{\partial s}=\beta(1-\beta^2)S \qquad (18)$$

The boundary conditions (6), (7), (8), (9) assume the form:

$$\text{for } x=1\begin{cases} y+\frac{2}{m^2-1}\frac{1}{\tau_0}\frac{\partial E}{\partial x}+\lambda^*\frac{\partial T^*}{\partial z}=S & (19)\\ y+\frac{1}{\tau_0}\frac{\partial y}{\partial x}-2(q-1)E=0 & (20)\end{cases}$$

$$\text{for } x=0\begin{cases} a-y=0 & (21)\\ a+y=2W \text{ or } \partial E/\partial s=0 & (22)\end{cases}$$

By (5) and (7) we have

$$S=We^{-\beta\tau_0 x} \qquad (23)$$

We have already seen in (2) that the quantity m can practically be taken to be constant; this is true even in a greater measure of the quantity N in which the numerator and denominator decrease simultaneously with the height. Let us assume that

$$m=\text{const},\ N=\text{const}$$

taking for m and N their mean Earth surface data.

Putting $T=280°$, $\alpha=7{,}25$ cm^2/g, $\rho_w=6.2\cdot10^{-6}$g/cm^3, $\rho=1.3\cdot10^{-3}$ g/cm^3, we obtain

$$N\approx10^5\omega$$

($1/\omega$ in seconds). Assuming that the characteristic time is 24 hours ($\omega\approx10^{-5}$), we stall find in our equation that $N\approx1$, and thus only the coefficient before $\partial E/\partial s$ in (18) will have the order of unity, whereas the coefficients before the derivatives of higher degrees will all be small ($m^2\approx3$, $\tau^0\approx10$, see (2)). This remarkable property of the equation (18) is characteristic of the problem of the boundary layer: it is only in the immediate vicinity of the two boundaries ($x=0$, $x=1$) that we shall have to solve (for small t) the full equation (18); everywhere else we shall have simply

$$N\frac{\partial E}{\partial s}\approx\beta(1-\beta^2)S\,*$$

This property disappears, however, if the characteristic time is, *e. g.*, 10 days or more, — the coefficients will then show a tendency to become gradually equal. The boundary layer of the Earth will extend in width, and eventually transformation of the air-mass will take place. These considerations accord well with the fact that the daily variations of temperature are largely confined within the limits of 1 km from the Earth surface, whereas the variations of temperature for the year are nearly invariable throughout the troposphere.

In principle, the solution of the equation (18) for arbitrary initial data and for the boundary conditions adopted by us does not present any difficulty. Here I content myself to consider an example of yearly variations of

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* Since $\beta\approx0$, we have practically $\partial E/\partial s\approx0$. In the general case this will reduce to the equality $c_p\rho\frac{\partial T}{\partial t}-\frac{c_p-c_v}{R}\frac{dp}{\partial t}\approx0$ which describes the well-known fact that in meteorological forecasts made 24 hours in advance the processes taking place in the middle troposphere can be regarded as adiabatic.

temperature. The solution of our equation (which is a linear one as are also the boundary conditions) can be written in the form

$$E = E_0(x) + \sum_1^\infty [E_{1n}(x)\cos ns + E_{2n}(x)\sin ns]$$

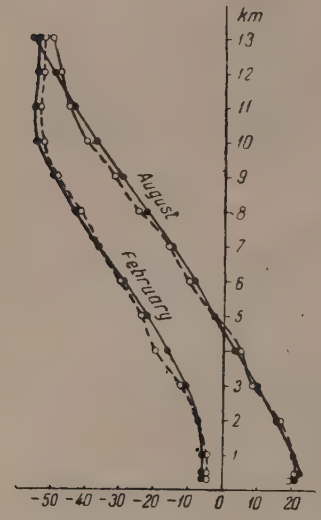
For E_0 (mean yearly distribution along the vertical) we can take the solution from ([2]). Substituting W as follows:

$$W = W_0 + \sum_1^\infty (W_{1n}\cos ns + W_{2n}\sin ns)$$

and denoting

$$E_n = E_{1n} - iE_{2n}, \qquad W_n = W_{1n} - iW_{2n}$$

we obtain an approximate solution of equation (18) for boundary conditions (19), (20), (21) and (22) in the form



Solid curve, actual variation of temperature; dotted curve, computed variation of temperature with height

$$E_n(x) \approx \left\{ \frac{m-\beta}{1-\beta}\frac{1}{m}\frac{\operatorname{ch}[\lambda_2(1-x)+\zeta]}{\operatorname{ch}(\lambda_2+\zeta)} - \frac{m-1}{m(1-\beta)}\beta e^{-\lambda_1 x} - e^{-\beta\tau_0 x}\frac{1-\beta^2}{2\beta}\frac{m^2-1}{m^2-\beta^2}\frac{W_n}{1-\frac{1-\beta^2}{m^2-\beta^2}\frac{m^2-1}{2\beta^2}nNi}\right\} \quad (24)$$

where

$$\lambda_1 \approx m\tau_0, \quad \lambda_2 \approx \sqrt{\frac{m^2-1}{2}\frac{Nni}{m^2}}\,\tau_0$$

$$\zeta = \frac{1}{2}\ln\frac{1+M}{1-M}, \quad M = \sqrt{\frac{m^2-1}{2m^2N}}\sqrt{\frac{\lambda^* c^* \rho^* \omega}{4\xi\sigma T^{*3}}}$$

As in ([2]), we have set $m = 1.75$, $\tau_0 = 12.6$, $\beta = 0.2$. The new quantities M and N are here: $M \approx 0.45$, $N \approx 0.024$, so that $\lambda_1 \approx 22$, $\lambda_2 \approx 0.8(1+i)$, $\zeta \approx 0.3$. The quantity W_n was taken according to Milankovich ([3]) for the latitude 40°. Assuming that the albedo is the same as in ([2]), we get

$$W_1 \approx 0.07i; \; W_2 = W_3 = \ldots = 0.$$

The results of computations for February and August are given in the adjoined figure. The figure also includes for the sake of comparison the mean four-year values of temperature for the same months as obtained at Omaha, in USA (sounding-balloon data for 1939—1942 ([4])). Less satisfactory is the agreement for spring (overestimated values of temperature) and autumn (underestimated values) months. But here, too, the divergence is comparatively small.

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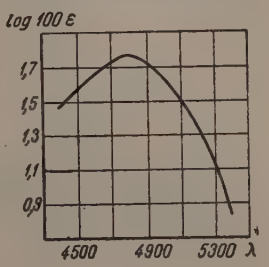
CHEMISTRY

ON DYES WITH ANTIPYRINE NUCLEI AND ON A SPECIAL CASE OF DEHYDROGENATION

By A. E. PORAY-KOSHITZ, Member of the Academy, B. A. PORAY-KOSHITZ and O. F. GINSBURG

The condensation products of aldehydes with two molecules of antipyrine (methylene-bis-antipyrine) and its products of substitution in the methylene group, described in literature (1) mainly as medicinals, must be referred from their structure to leuco-compounds of dyes. The latter, being analogs of triarylmethane dyes or of the simplest cyanine dyes, may be capable of functioning as optical sensitizers.

Actually, benzylidene-bis-antipyrine, prepared according to Knorr (1) from benzaldehyde and antipyrine, yielded after oxidation a brick-red dye that crystallized beautifully as a nitrate and had a light absorption maximum at 4800 Å. This product rendered photographic plates sensitive to green and yellow-green rays.



By its optical properties it was very similar to pinoflavole.

With the aim of increasing the depth of the colour of the dyestuff, the corresponding leuco-compound was synthetized from 2 moles of antipyrine and 1 mole of dimethyl-*p*-aminobenzaldehyde. The dihydrochloride of this compound possessed the interesting property, as far as known by the authors not observed previously, of splitting off a molecule of hydrogen when heated without any catalysts to 120—140° C, passing over into a red dyestuff with a melting point 178°. The optical properties of this dye have not been investigated as yet. The work is being continued.

Experimental. Benzylidene-bis-antipyrine hydrochloride was prepared according to Knorr. Yield 82—84.5 per cent m. p. 152° (compare (2)). The free base was isolated through the action of an alkali and proved to be colourless, m. p. 206°. The dye was obtained through the oxidation of a suspension of the sulphate in HCl with nitric acid (sp. weight 1.4) under reflux. The dark red solution was made alkaline with NH_3. The colourless solution of the carbinol base in NH_3, after the separation of a resinous precipitate, was acidified with NHO_3.

There separated red crystals of the nitrate of the dye. M. p. 245°. Yield up to 34.2 per cent of the theoretical.

Analysis: 1) subst. 0.1078 g: N 13.0 cm³, $p = 760$, $t = 20°$, N 13.61 p. c.
2) » 0.0968 g: N 11.6 cm³, $p = 759$; $t = 22°$, N 13.67 p. c.
Calculated for $C_{29}H_{27}N_5O_5$ N 13.36 p. c.
3) subst. 0.1032 g: 0.2502 g CO_2, 0.0481 g H_2O; C 66.1 p. c., H 5.19 p. c.
4) » 0.1123 g: 0.2738 g CO_2, 0.0517 g H_2O; C 65.97 p. c., H 5.11 p. c.
Calculated C 66.25 p. c., H 5.17 p. c.

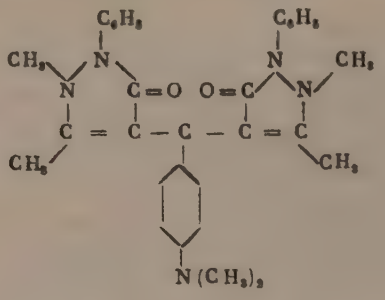
The absorption spectrum curve is given in the figure. It has a maximum at 4800 Å.

The photograhic plate was immersed for 3 min. in a 1 : 50 000 alcohol-aqueous solution of the dye. The colour circle was photographed and sensitivity to yellow-green and green rays was noted. The substance dyes animal fibres and tanned cotton into a brick-red colour. A dyed sample was exposed during 20 days to the action of daylight. It faded but slightly, the shade remaining unchanged.

The base of the dyestuff was isolated from the nitrate through the action of NaOH.

It is a light brown powder (from alcohol) that decomposed about 140°.

Dimethyl-*p*-aminobenzylidene-bis-antipyrine dihydrochloride was prepared by mixing solutions of antipyrine and dimethyl-*p*-aminobenzaldehyde in HCl, sp. weight 1.19, and by diluting with water.



Pink crystals. After three precipitations with water from HCl, sp. weight 1.19,—colourless crystals. Yield 66.5—75 per cent of the theoretical. When the melting point is determined both in an open and in a sealed tube at 120—140° the substance swells, becomes red, then hardens and melts again at 178°.

Analysis: Weight portions 0.2964 and 0.2694 g; AgCl 0.1426 and 0.1288 g
Found Cl 11.92 p. c., 11.83 p. c.
Calculated for $C_{31}H_{33}N_4O_2$ Cl 12.2 p. c.

Evolution of Hydrogen. The dihydrochloride was heated in a current of CO_2 for 1 hour at 175—180° (in a sulphuric acid bath). A red substance formed in the reaction tube. The evolving gas was collected in an eudiometer over a 50 per cent KOH solution.

Combustion in a Hempel apparatus in the presence of platinized asbestos proved the gas to be pure hydrogen.

1. From 0.4050 g of the substance 13 cm³ of hydrogen were obtained. Theoretical amount of H_2 15.6 cm³.

2. From 0.3989 g of the substance 12.3 cm³ of hydrogen were obtained. Theoretical amount of H_2 15.4 cm³.

The leuco-base of the dyestuff (dimethyl-*p*-aminobenzylidene-bis-antipyrine) was isolated from the solution of its dihydrochloride with sodium hydroxide. After crystallization from aqueous alcohol with the addition of a small amount of sodium carbonate (otherwise the solution becomes pinkish) its melting point was 197—198° (with decomposition). It is soluble in alcohol, benzene, ether.

Molecular weight determination:

Benzene 6.753 g, substance 0.1284 and 0.1255 g.
Depression 0.18 and 0.185°. Found: *M* 516 and 491; average *M* 503.5.
Calculated for $C_{31}H_{33}N_5O_2$ *M* 507.

Analysis: Weight portions 0.1128 and 0.1043 g; N 14.0 cm³, $p=755$, $t=19°$;
N 12.8 cm³, $p=758$, $t=20°$.
Found N 14.12 and 13.98 p. c. Calculated 13. 8 p. c.

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PHYSICAL CHEMISTRY

PHENOMENON OF THE LOWER LIMIT IN THE LOW-TEMPERATURE POLYMERIZATION OF HYDROCARBONS

By S. E. BRESLER

(Communicated by A. F. Joffé, Member of the Academy, 8. IX. 1944)

A unique reaction that has claimed the attention of chemists in connexion with the process of obtaining high molecular polyisobutylene is the low-temperature polymerization of hydrocarbons. This reaction takes place in liquid hydrocarbons at fairly low temperatures, of the order of —80 to —100°C, under the action of highly active catalysts such as the boron fluoride BF_3. Since the catalyst dissolves in hydrocarbons, this is an instance of homogeneous catalysis.

The whole picture of the polymerization process is most unique. Notwithstanding the low temperature, the reaction is extremely violent, accelerating of itself, and ends in a closed volume in a fraction of a second. As it proceeds, a large quantity of heat is evolved, which causes frequent quite sizeable pressure increases and bursting of apparatus. The auto-acceleration of the reaction, as experiment showed, is not thermal in nature. As a matter of fact, if the polymerization is carried out in solutions in which the solvents are low-boiling hydrocarbons, such as ethylene (b. p. —104°C), ethane (b. p. —78°C), *etc.*, isotherma conditions are maintained in the reaction mixture. However, in this case the characteristic course of the process is exactly the same, as when there is no solvent. Only the structure of the resulting polymer is essentially affected by the introduction of the solvent. For example, polymerization of isobutylene in low-boiling solutions under conditions that ensure the carrying out of the process at a low temperature, yields rubber with extremely high molecular weight (M reaching the order of 250 000), and this notwithstanding the explosive-like course of the reaction.

On the basis of these results it could be assumed that the auto-acceleration of the reaction is of a chain-like nature, and that in these reactions we are dealing with unique explosions generated at —100°C. Extending our observations to the polymerization of divinyl and isoprene, we discovered the phenomenon of a critical initial temperature, the exact equivalent of the lower limit in chain explosions.

Here we shall pause to discuss in brief the experimental method employed. The apparatus in which the reaction took place is shown in the accompanying figure. It was submerged in a cooling mixture of acetone and methanol, cooled to the required temperature by liquid air. Then the hydrocarbon in the gaseous state was admitted, and after it had condensed, boron fluoride was introduced through the tube B from a mercury gasometer. When the reaction was over, the polymer was purified of residual active products by washing with liquid ammonia, which was condensed with the aid of the Dewar cap A. Experiment showed that if the polymer is not washed, secondary processes set in when it is warmed up to room temperature, which lead to depolymerization and breakdown of the polymer. Upon the completion of an experiment the apparatus could be broken,

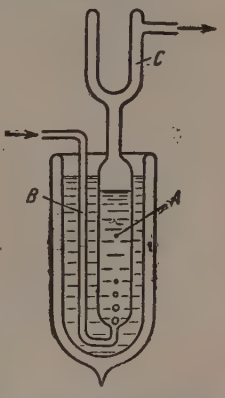
and the finished polymer removed from it. This simple method is a most convenient one for qualitative experimentation, since the entire process takes place in full view of the investigator.

New is the following phenomenon we observed. Divinyl, in which up to 0.3 per cent of boron trifluoride is dissolved, practically does not polymerize at all when left standing for many hours, until the temperature is raised to —76°C. Then an auto-accelerating reaction suddenly sets in, that attains the explosive-like proportions mentioned above. The same holds for isoprene, except that in this case the critical temperature is somewhat higher, namely, —68°C.

It is known that when $f-g>0$ in the equation

$$dn/dt = n_0 + (f-g)\,n$$

where n_0 is the number of centres originating per unit of time, f is the chain branching constant, and g is the centre perishing constant, chain auto-acceleration of the reaction occurs. If $f-g$ is very small, such a reaction is practically undetectable. On the other hand, when the probability of branching ceases to be compensated by the probability of perishing of the centres, the progress of the reaction by stages becomes impossible, and auto-acceleration sets in. The condition for self-ignition includes as a critical quantity the branching constant f, which changes with the temperature according to the Arrhenius rule. But in addition, in our case the number f depends upon the amount of the catalyst introduced. At a given constant temperature it should be expected that the auto-accelerating reaction becomes critical, jerky, when the concentration of the catalyst reaches a definite value. Experiment fully confirms this assumption. It was found that no less than 0.05—0.1 per cent by weight of the catalyst has to be added to the liquid hydrocarbon to initiate the reaction. It should be noted that the experimental value of this concentration limit is poorly reproducible, since it is markedly affected by the slightest impurities in the reagents. This is natural, since at low temperatures the complexes that BF_3 forms with most polar organic molecules are found to be extremely stable. As for the polymers obtained from dienes under the action of BF_3, these are insoluble resin with volume-branched chains; evidently, both double bonds of the diene are activated and react in the polymerization process.



Apparatus for with boronr trifluoride polymerizationw

Leningrad Physical-Technical Institute.
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PHYSICAL CHEMISTRY

KINETICS OF CATALYTIC PROCESSES ON HIGHLY NON-UNIFORM SURFACES

By S. Z. ROGINSKY, Corresponding Member of the Academy

The control band method developed by the author in recent works [1,2] makes it possible to elucidate the general principles of the kinetics of contact reactions on non-uniform surfaces, which are characterized by a broad range of values of the kinetic and adsorption coefficients.

Typical contact reactions are complex processes with many stages. At least one of the successive stages before final dynamic equilibrium is established between the initial reactants and the resultants, is incomplete. The reactants for this stage are in complete equilibrium with all of the preceding substances, but with none of the following ones. The equation for semi-equilibrium of this kind makes it possible to connect with the initial reactants the concentration of the intermediate substances figuring in the irreversible stage. It is this equilibrium in conjunction with the law of the kinetics and the absolute rate of the incompletely reversible stage that determines the fundamental kinetic peculiarities of the observed overall reaction (Fig. 1).

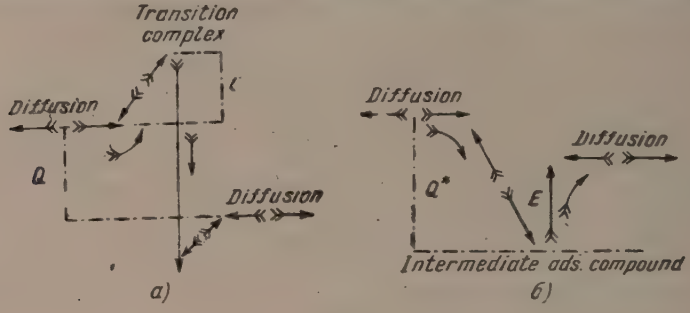

Fig. 1

The rate constant of most catalytic reactions increases exponentially with temperature, owing to the presence of an endothermal stage that may be either completely reversible or irreversible. In the first case there is an equilibrium number of reversible endothermal surface formations in the system, which undergoes further transformation with a certain frequency ν. In the second there are intermediate equilibrium exothermal surface formations in the system, which undergo further transformation at velocity whose rate constant contains the factor $\exp(-E/RT)$, where E is the energy of an incompletely reversible endother-

mal stage. This difference in sign of the heat effect of formation of an intermediate product is of decisive significance for the kinetics.

Let us assume for simplicity that the same law of the kinetics of the reaction applies to the entire surface, and that from the kinetic standpoint every site of the complex surface acts independently of the others, and, moreover, that definite distributions exist with respect to the activation energies of the catalytic process $\rho(E)$ and with respect to the heats of adsorption of the initial reactants and the resultants: $\rho(Q_1)$, $\rho(Q_2)$, $\rho(Q_3)$, or with respect to Q^*, *i. e.* the heat of formation of the exothermic intermediate product. Let us assume further that the variations in the factor before the exponential term, k_0, in the equation $k = k_0 \exp(-E/RT)$ are of secondary importance for the rate constant of the process, which is not always true. Then it is easy to show that the sites with activation energies lying within a very narrow interval of energy of activation $\sim RT$ bear the brunt of the burden.

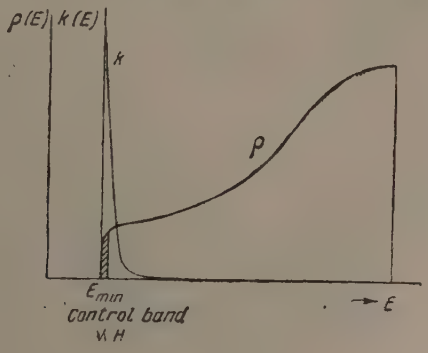

Fig. 2

Suppose that the distribution of the sites with respect to activation energies is like that shown in Fig. 2. The character of the process is determined by the sign of the heat effect of formation of an intermediate state and the shape of the dependence of E upon Q^*. There are four possible cases.

1. The control band lies to the left for minimum values of E. The intermediate substance that figures in the semi-equilibrium is endothermal and there is no self-poisoning by the products, owing to the weak adsorption in the zone of the control band.

Then the rate of the process will be expressed by the equation

$$w = K \Pi p_i \qquad (1)$$

where

$$K = \int_{k_2}^{k_1} \rho(k)\, k dk \text{ and } \Pi p_i = p_A^{\nu_A} p_B^{\nu_B} \qquad (1b)$$

ν_A, ν_B . . . being stoichiometric coefficients.

Integration of (1b) gives a value of K equal to $K_0 \exp(-E/RT)$, where

$$K_0 = k_{01} RT \rho_{k_1} \qquad (1c)$$

No matter what the exact composition of the endothermal transition product, the fraction of the surface it covers cannot be in the least significant.

Let us denote by Ψ the unoccupied fraction of the total surface. Then the rate of the process will be

$$w = \Psi K' \Pi p_i \qquad (2)$$

So long as the surface is not poisoned to any considerable extent by the reaction products, the reaction should proceed at a rate proportional to some integral or simple fractional power of the partial pressure, retaining these characteristics in an arbitrarily wide range of absolute pressures. The process will fit the Van't Hoff equations of homogeneous kinetics.

The character of the transition complex may be established from the power of the partial pressures of the initial reagents in equation (2). The simplest type of reactions exhibiting quasi-Van't Hoff kinetics of this nature, which resemble the Langmuir kinetics at very low coverings, are those limited by the activated adsorption of one of the initial reactants. The temperature dependence is normal and gives the true value of E for the sites of the control band. Reactions of this type are evidently very widespread at high T.

2. In those cases where the catalytic reaction is accompanied by a decrease in activity of the most active sites, whether due to E_{cat} and Q_{ads} of the reaction

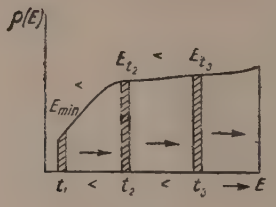

Fig. 3

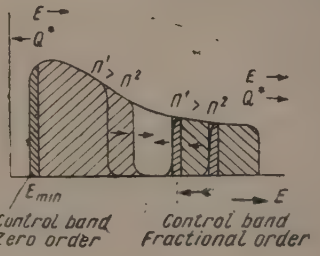

Fig. 4

products varying in the opposite sense, or to spoilage of the surface by the catalytic reaction, *etc.*, the control band begins to move, shifting in the direction of increasing values of E (Fig. 3). Due to this the activation energy ceases to be a constant and changes continually as the reaction proceeds. In the case of spoilage of the contact agent by the reaction products, the kinetics of the process is hardly distinguishable from the kinetics of activated adsorption.

The change in E with time is described by the kinetic isotherm equation

$$E = RT \ln \bar{k}_0 t \qquad (3)$$

except that here $\bar{k}_0$ may be only some β-th part of k_0 for the catalytic process. For different types of distribution diverse equations are obtained for the kinetic isotherms and isochrones ([2]). Thus, for uniform distribution $q = a \ln t + C$ and for exponential distribution $q = at^{1/n} + C$.

Examples of reactions fitting such equations are the low-temperature oxidation of carbon monoxide on manganese peroxide and the catalytic hydrogenation of ethylene ([3],[4]).

In those cases where the self-poisoning by the products is reversible, individual differences among surfaces with a different type of distribution vanish. When the initial concentrations are constant and the concentration of the poisoning product increases as the process proceeds, the most diverse types of $\rho(E)$ functions lead to $q \sim t^{1/n}$. The only condition is a linear relation between the activation energy of a site and the heat of adsorption of the poison on this site: $E = E_0 - \beta Q$.

3. Of an entirely different character are the kinetics of catalytic reactions when the equilibrium stage preceding the irreversible stage is exothermal. The latter may be, for example, the desorption of a firmly adsorbed product, or the decomposition of the exothermal surface compound. When the transition complex is of such a nature, the character of the functional dependence between the heat of formation of the transition complex Q^* and the energy E of the endothermal process, in which this transition complex enters, is of decisive importance (Fig. 4).

When these two energies vary in the opposite sense, the stationary control band lies at the edge for minimum E and maximum Q^*. In the most characteristic case all sites are completely occupied by molecules of the exothermic formation from E_{min} to $E_{boundary}$, located at a considerable distance from the edge (Fig. 4, left).

Variations in the total pressure or in the composition of the gas will shift $E_{boundary}$, changing nothing within the control band zone; therefore, so long as poisoning by the reaction products is absent, the reaction should take place at a constant velocity

$$E = E_{min}, \quad \psi = \text{const}, \quad w = k = \text{const} \tag{4}$$

and be of the zero order for all substances that go to make up the transition complex (which does not exclude the retarding action of the substances that do not enter into its composition). These characteristics should be quite stable. Cases of a zero order in two components were actually observed in the catalytic hydrogenation of a number of compounds ([5]). In our treatment it is not imperative to employ the conception of two types of qualitatively different sites, on each of which only one component may be adsorbed*.

Self-poisoning starting with the control band should set the latter in motion and cause the appearance of kinetics characteristic for activated adsorption, but with a velocity independent of the initial concentrations of the substances entering into the composition of the complex.

4. Quite a different picture is obtained when E and Q^* vary in the same sense. In this case to the left of E_{max} up to a certain value of $E_{boundary}$ the total surface is completely occupied, and the contribution of sites of a given kind to the total velocity depends upon $k—k_0 \exp(—E/RT)$ and upon the value of ρ at a given E.

Therefore, there are two possible positions of the control band: at the edge of the occupied part of the distribution $\rho(E)$ in the case where E changes more slowly than Q^*, and to the left at the edge of the distribution when E changes faster than Q^*. In the second case the kinetics is hardly distinguishable from the kinetics for the case where the transition complex is endothermal, the energy of activation is constant and the dependence of the velocity upon the concentration conforms to Van't Hoff's equation. In the first case the position of E depends upon the value of Q^*, while this latter may be approximately found from the following equation:

$$Q^*_{boundary} = Q^*_{0.5} = -RT \ln \left(\frac{\Pi p_A}{b_0}\right) \tag{5}$$

where Πp_A is the product of the partial pressures of the reacting gases raised to powers corresponding to their stoichiometric coefficients (Fig. 4, right). Then:

$$E = E_0 + \beta Q^* = E_0 - \beta RT \ln \left(\frac{\Pi p_A}{b_0}\right) \tag{6}$$

Hence the value of the overall velocity w, coinciding with the reaction rate on the control group of the sites, will be expressed by the equation:

$$w = A_0 \exp(-\gamma E/RT) = K[\Pi p_A]^m \tag{7}$$

where $m = \gamma\beta$, $\gamma = 1$ for uniform and power distributions, and $\gamma = 1 - \alpha RT$ for exponential distributions with the coefficient α in the exponent.

Inspite of the movement of the control band, the energy of activation, determined by the thermal coefficients of the velocity, remains constant, and equal to γE_{min}. The concentrations of all the substances in the transition complex enter into the equation with fractional exponents.

Characteristic is the fact that the ratio of the exponents for individual concentrations is equal to the ratio of the stoichiometric coefficients of the latter in the transition complex.

* Though such an interpretation is supported in the work by A. A. Ba'andin (report to the Chemical Session of the Academy of Sciences of the USSR, 12.X.1944).

Kinetics of this kind is characteristic for ammonia synthesis ([6]) and for the contact oxidation of sulphur dioxide ([7]). It is not connected, as was previously assumed, with a definite type of distribution, and follows from the width of distribution and the fact that the relation between E and Q^* is linear and varies in the same sense.

Equation (7) is an approximate equation, since in its derivation the slight dependence of A_0 upon E was never taken into account, whereas this factor is actually constant for all sites only in the case of uniform distribution.

Thus, for a surface with a broad distribution of sites with respect to the heat of formation of the transition complex, which may or may not coincide with the energy of activation, the Langmuir system of equations of the kinetics is replaced by a new system which includes: 1) quasi-Van't Hoff kinetics equations; 2) zero orders for one or several components; 3) logarithmic or fractional-power increase of the quantity adsorbed with time in the case of Van't Hoff dependence or complete independence of the concentrations of the initial reactants; and finally, 4) fractional powers of the concentrations of the reaction components.

The first two types represent in an exaggerated form the extreme cases of very small and very large coverings in the Langmuir kinetics; the fourth also resembles the Langmuir kinetics for the case of medium coverings. However, the difference between similar cases in the kinetics for uniform and highly non-uniform surfaces is great.

Primarily it manifests itself in the anomalous stability of all three types, which is not characteristic of uniform surfaces. In the case of uniform surfaces the appearance of the third type of self-inhibition cannot be brought under the scope of the laws of the kinetics of activated adsorption unless special assumptions are made, such as the existence of a distorting effect due to the repulsive forces.

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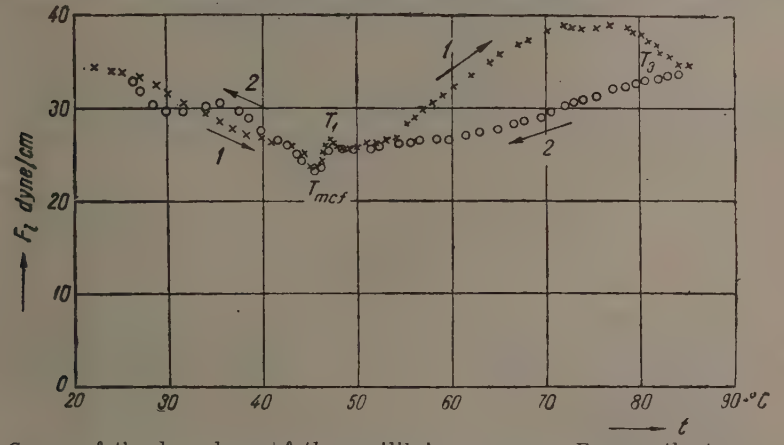
PHYSICAL CHEMISTRY

EQUILIBRIUM BETWEEN TWO- AND THREE-DIMENSIONAL HYDRATES OF THE HIGHER ALCOHOLS AND NEW PHASE TRANSFORMATIONS

By A. A. TRAPEZNIKOV

(Communicated by A. N. Frumkin, Member of the Academy, 20. II. 1945)

In a recent work [1] it was discovered by the author that crystals of the higher aliphatic compounds (fatty acids, alcohols) interact with water to form hydrates which differ essentially in their properties from the substances in the unhydrated state. A new method of studying the bulk hydrates was described, based



Curves of the dependence of the equilibrium pressure F_e upon the temperature t for a monolayer of ceryl alcohol $C_{26}H_{53}OH$ in equilibrium with small crystals on 00.1 N NACl

on the measurement of the two-dimensional pressure F_e of the monolayer in equilibrium with the bulk phase (crystallohydrate) as a function of the temperature t.

Using tetradecyl alcohol as an illustration, a new type of curve $F_e = f(t)$ was given, characterizing the equilibrium between the monolayer and the three-dimensional hydrates.

Research into the equilibrium between the three-dimensional phase and the monolayer for the highest homologues of aliphatic series of alcohols and fatty acids has led to the discovery of new, extremely interesting phase transformations, both in the bulk hydrates and in the monolayers.

The latent heats of the phase transformations have been computed on the basis of the $F_e = f(t)$ curves, which permits the problem of structure and state both for three- and two-dimensional hydrates to be approached from a new angle.

Until now the conformity between the structure and the properties of two-dimensional systems (monolayers) and three-dimensional systems has not been established.

Anhydrous crystals are often misinterpreted as analogues of condensed monolayers. The author has ventured the opinion [1] that such an analogy may be made only with respect to a three-dimensional phase in the form of hydrates.

Now this is confirmed on the basis of a comparison of the latent heats as derived from $F_e = f(t)$ curves and the temperatures of the phase transformations in the monolayer and the bulk hydrate.

Curves *1* and *2* for ceryl alcohol (see figure) were obtained for repeated heating and cooling, respectively. At temperatures below 50 °C they practically completely coincide, and hence represent thermodynamic equilibrium changes in state. The right part of curve *1* apparently represents the formation of a non-equilibrium hydrate structure.

There are three characteristic points on these curves: T_{mcf}, T_1 and T_3 which correspond to discontinuities in the latter, and hence represent some changes or other in state of the system. If the formation of the monolayer from the crystal is regarded as two-dimensional solubility or evaporation, one may, as was done by Cary and Rideal [2] for the first time, employ the Clapeyron-Clausius equation $\frac{dF_e}{dT} = \frac{Q}{T(A_2 - A_1)}$ to calculate the latent heats Q of transition of the molecules from the three-dimensional phase to the monolayer. Here T is the absolute temperature, and A_1 is the area occupied by a mole of the substance in the bulk state, which is sufficiently small to be disregarded in comparison with the area A_2 occupied by a grammol of the substance in the monolayer (area per molecule a is roughly taken to be equal to 20Å^2).

Such a computation for ceryl alcohol reveals that at the point $T_{mcf} = 45.5°$ C, which we have called the melting temperature of the crystalline film, phase transformation occurs accompanied by the absorption of approximately 30 000 cal./mole. Phase transformation (opposite in sign) seems likewise to occur at the point $T_1 = 47°$ C, which in this case should be evidently ascribed to the bulk phase. During this transformation also there is absorption of energy of the order of 30 000 cal./mole. At the point $T_3 = 80°$ C the bulk crystal is transformed into a turbid anisotropic liquid, *i. e.* this temperature corresponds to a visible melting of the crystallohydrate*. At this point the latent heat effect (as indicated by the cooling curve) is only about 3000 cal./mole, *i. e.* one-tenth of that at T_1. The respective figure obtained from the heating curve is not large either, not above 8000 cal./mole.

According to direct calorimetric measurements made by Garner [3], the latent heat of crystallization for crystals of anhydrous long-chain aliphatic compounds (fatty acids, paraffins) amounts to approximately 1 000 cal. per a mole of CH_2 groups in the hydrocarbon chain, which would give about 26 000 cal./mole for ceryl alcohol (unfortunately, direct measurements have not been made with ceryl alcohol). It is apparent that the latent heat effects that we have detected at T_1 and T_{mcf} are very close to this quantity, whereas the heat effect in the case of visible melting at T_3 is much lower.

On the basis of this comparison it follows that hydration of the three-dimensional phase very essentially changes the state and structure of the three-dimensional crystal. Thus, instead of 26 000 cal./mole being expended to fuse the crystal with rupture of the bonds between the CH_2 groups of adjacent molecules, a total of only 3 000 cal. is required in the hydrated crystal. On the other hand, the full amount of energy needed to rupture these bonds is actually expended in the hydrated state too, although at a temperature that is much lower (33° lower for ceryl alcohol) than the temperature of visible melting. Hence, the molecules of water wedge inbetween the molecules of the alcohol in the crystal, forcefully

* The point T_2 which corresponds to intermediate transformation is absent from the cooling curve, and its discussion here is omitted for the sake of brevity.

pushing apart the hydrocarbon chains (at $t>T_1$). From the order of magnitude of the heat effect, the phase transformation at T_1 appears to be a melting. But as a matter of fact, it is probably a case of polymorphous transformation of crystal structures. In this event, it differs essentially, so far as energy is concerned from the transformation of the β-modification into the α-modification in anhydrous crystals.

Highly interesting is the fact that for ceryl alcohol the latent heats of transformation of the hydrated crystal at T_1 and of the monolayer at T_{mcf} are practically identical.

This shows that transformations of the initial type involving the destruction of the very same bonds between the CH_2 groups of parallel hydrocarbon chains, take place in both states.

This resemblance shows that hydration plays a most important rôle both in the structure of the monolayers, even of the most condensed-solid monolayers, and in the structure of the bulk phase, which as a result of hydration thus approaches in properties to the two-dimensional state.

In the monolayer this transformation consists in melting, which is proved by quantitative measurements of the mechanical properties of the monolayer by the distorsion and oscillation method (4). It is found that when $t<T_{mcf}$ the monolayer is elastic and strong, and when $t>T_{mcf}$ the monolayer is highly viscous (of an anomalously viscous structure).

In view of the abrupt jump-like qualitative change in the mechanical properties of the monolayer at the point T_{mcf}, accompanied by a sizeable heat effect, we maintain that when $t<T_{mcf}$ the monolayer is solid-crystalline, and when $t>T_{mfc}$ the monolayer is liquid-crystalline (anisotropically liquid). The conception of a liquid-crystalline state differing from a solid-crystalline state is most essential, it is characterized by definite mechanical properties—anomalous high viscosity and the absence of evident rigidity. This state was previously regarded by nearly all investigators as a «plastic-solid» state. It undoubtedly differs from the true-liquid isotropic state as well, since there is a quite definite temperature $T_{ml-cf}>T_{mcf}$ at which the viscosity of the monolayer, in a state of equilibrium with the three-dimensional phase, can practically no longer be measured. At this point T_{ml-cf} transition of the condensed monolayer into a liquid-expanded state (isotropically liquid) takes place.

The described transformation resembling ordinary melting that takes place in the monolayer at the point T_{mcf} had not been detected previously. Cary and Rideal looked for the transition from the solid state into the liquid state in a monolayer of myristic acid in equilibrium with the crystal, but without success. Now this may be explained by the fact that the monolayer of myristic acid is no longer solid at room temperature, even under equilibrium pressure.

In the case of acids, monolayer melting analogous to that described for ceryl alcohol may be detected only in the higher homologues, the C_{20}—C_{26} acids, for example.

Monolayers of the C_{14}—C_{18} alcohol and acid homologues are already found to be liquid-crystalline at ordinary temperatures, and require only a very small latent heat to pass over into the isotropically liquid (liquid-expanded) state.

The detected melting of the monolayer and of a series of new two-dimensional structures opens up a new way for studying the state of fully saturated monolayers, as related to temperature, as well as of unsaturated ones along the $F=f(a)$ curve obtained upon changing the area per molecule a by the compression method. Thus, it is found that the point at which the $F=f(a)$ curve becomes discontinuous, which was previously regarded as the point of solidification, is not that at all. The monolayer is solid along the entire $F=f(a)$ curve when $t<T_{mcf}$, and liquid-crystalline along the entire $F=f(a)$ curve when $t>T_{mcf}$.

It must be noted that the two-dimensional phase in the monolayer of C_{18}-alcohol described as S-phase by Harkins and Copeland (5) should, according to our data, be regarded as liquid-crystalline, which appears when $t>T_{mcf}$. It is equivalent to the liquid-crystalline phase of ceryl alcohol, which is likewise cha-

racterized by a decrease in viscosity with temperature in the temperature range from 45.5 to 49° C. From $t=49°$ C the viscosity of the equilibrium monolayer again rapidly increases, which is connected with the formation of another liquid-crystalline structure that Harkins and Copeland (5) call a new «superliquid».

The fact that for $t>T_{m.f}$ the curve, bringing out the variation of monolayers in the liquid-crystalline state with increasing temperature, runs through a minimum, is a common property in different classes of substances: alcohols, acids, triglycerides.

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GEOLOGY

ON THE THEORY OF SALT TECTONICS

By N. M. STRAKHOV

(Communicated by V. A. Obruchev, Member of the Academy, 29. VII. 1944)

In the literature dealing with salt tectonics, this term is applied to structures produced by intraformational displacements of salt alone, without participation of any other member of the formation, anhydrites and clays in particular.

Some of the authors attach great importance to the processes of hydration of anhydrites and to the resulting increase in their volume.

In spite of the fact that this way of interpreting salt tectonics has become nearly a current one, it is far from being duly backed by factual material. A tectonic analysis of salt structures has recently been carried out in such regions (Persia, Gulf Coast area in the United States, Ural-Emba region in the USSR) where the halogen formations are but partly exposed by natural outcrops or holes, and where the behaviour of the different members of halogen formations is a matter of guess-work rather than of sound judgment based upon precise data.

The territory of the Ishimbaevo area of the Near-Ural region where the author was working in 1941—43, is very interesting in this respect, since a vast number of holes (over 800) has been sunk there through the whole of the halogen formation, which made it possible directly to investigate the «internal anatomy» of the salt structures.

To be able to follow the idea developed below, it should be borne in mind that the Lower Permian (Kungurian) halogen formation of the Ishimbaevo area of the Near-Ural region is composed of three horizons: the lower or salt-bearing one, composed of anhydrites and halitites; the middle or gypsum-anhydrite one, formed only of sulphate rocks (gypsum, anhydrite); and the upper one (passage beds), consisting of sulphatized marls, clays, and sandstones.

When examining the structural map of the surface of the halogen formation (*i. e.* of the surface of the passage beds), it was found to be very complicated. Several high ridge-like bulges can clearly be distinguished (Allaguvatovo-Pokrovsk, Kusiapkulovo-Ishimbaevo, Smakaev, Baigusin, Termen-Elga, Mashash) separated by deep trough-like downwarpings (Kashkana, Kazhak, Smakaev, Kuzminov, Baikov troughs). The absolute marks of the roof of the formation of the bulges are generally above zero, reaching +126 and even +170 m; in the central parts of the troughs they drop down to —318 and —365 m in the North, and to—751 and—851 m in the South. The range of variation in height and in the nature of the bulges and depressions vary from North to South. In the North these elements of the relief of the roof of the formation are less clearly pronounced, smoother in their outlines, with gradual transitions from elevations to depressions; diapir piercements on the bulges are absent. In the South, the range of variation of the relief increases sharply, the bulges become more steep-walled, while their arc is complicated by diapir piercements of gypsum-anhydrite rocks.

It is essential to note that when viewed from a purely geographical aspect, there is no resemblance whatever between the outlines of the roof of the halogen formation and its basement; on the contrary, a distinctly pronounced disharmony is observed.

In order to interprete the origin of this disharmony, a systematic study was carried out of the total thickness of rock salt corresponding to the areas of bulges and troughs of the halogen formation. The results of their investigation proved of great interest. In the area of the Kusiapkulovo-Ishimbaevo bulge, the thicknesses of the salt range from 288 to 452 m; on the Smakaev elevation, from 546 to 602 m; on the Termen-Elga elevation, from 386 to over 852 (approaching more closely the latter than the former value); on the Yuldash elevation, from 610 to 745 m. In the intervening depressions between the bulges, the picture is quite different. In the centre of the Kazhak depression the holes failed to reveal the presence of salt at all, and along the periphery of the trough the thicknesses of the salt were found to attain 199 and 273 m only (holes 2/31, 1/31). In the central part of the Kuzminov depression hole 1/29 yielded only 105 m of salt; and hole 2/29, about 220 m. In the Ishimbaevo depression some holes did not produce any salt at all; one—96 m, and only one single hole 7/5 (along the periphery of the trough) opened up 566 m of salt. In the Kashkara trough in the centre no salt at all was found; along the periphery its thickness was up to 95 m; in the Yarbishkadak trough one single hole yielded 30 m of salt. From these data it is quite obvious that with the depressions of the Kungurian roof coincide the minima of salt thicknesses, or else salt is altogether absent, while with the bulges of this structural surface is associated a distinct maximum of rock salt. As such a distribution of salts does in no way reflect the peculiarities of the Artinskian basement, it should be considered merely as a secondary phenomenon, caused by redistribution of the plastic masses inside the Kungurian deposits in the course of post-Kungurian processes. The points of squeezing-out (removal) of salts have naturally produced depressions; and those of secondary accumulations of salts, bulges.

But is the configuration of the Kungurian roof actually determined by deformation and migration of salts only? An answer to this question is given by the study of the gypsum-anhydrite horizon, overlying the salt-bearing horizon. It has been found that on the Pokrovsk-Allaguvatovo bulge the thickness of this horizon ranges from 200 to 340 m; on the Kusiapkulovo-Ishimbaevo, from 290 to 365 m; on the Termen-Elga, from 250 to 425 m; on the Smakaev, from 245 to 545 m; on the Baigusin, about 285 m; on the Yuldashev, from 285 to 370 m. In other words, on the bulges the thickness of the gypsum-anhydrite horizon usually exceeds 250 m, amounting to 400—500 m. On the limits of the bulges in the zones transitionary to troughs, the values mostly range between 150 to 250 m, but in a few cases also very low thicknesses, of some tens of meters, are recorded (*e. g.*, hole 1/85—85 m, hole 21/3—80 m, hole 3/14—71 m, *etc.*). As to the troughs, the thicknesses of the gypsum-anhydrite horizon attain there 100—150 m, sometimes dropping below this value (the Kashkara trough, 34 to 150 m; the Kazhak, 100 to 161 m; the Kuzminov, 70 to 135 m, *etc.*). Thus, the thicknesses of the gypsum-anhydrite horizon repeat in a striking manner the picture of the distribution of thicknesses of rock salt. In either case the maximum accumulations are associated with the areas of bulges, while the minimum thicknesses correspond to the areas of troughs.

In interpreting the origin of this peculiar picture of localization of the sulphate horizon according to salt structures, it should be borne in mind that the gypsum entering into the composition of the horizon is a secondary one, owing its origin to the hydration of anhydrites, which is evidenced by its being present on bulges only (*i. e.* near the day surface), while it is absent in troughs (*i. e.* far from the surface). The hydration of anhydrites being accompanied by an increase in volume, this might be taken to account for the fact that the total thickness of the gypsum-anhydrite horizon over the bulges is abnormally great. An estimate shows, however, that this conclusion is wrong. Knowing the total thick-

ness of the gypsum over the bulges, it was possible to think that the hydration of anhydrites —if it is taken for granted that the whole increase in volume (57 to 64 per cent) takes place only in the vertical direction—could cause an increase in the thickness of the gypsum-anhydrite horizon by no more than 50 to 80 m, but as a rule much below this value. As a matter of fact, however, this thickness of horizon in the troughs is always less than 150 m and frequently less than 100 m, while on bulges it usually exceeds 250 m, and is frequently over 300—400 m.

It is far from being proved, moreover, that the increase in volume upon hydration is always equal to the theoretical 57—64 per cent; neither is it certain that the increase in volume always proceeds in the vertical direction only. Consequently, the increase in the thickness of the gypsum-anhydrite horizon on swells is essentially a tectonic phenomenon, related to a displacement of anhydrites from under the troughs of the roof of the formation towards the bulges. This fundamental process has been overlapped, in a slight and indefinite measure, by the influence of anhydrite hydration.

It is clear that this kind of intraformational displacement of anhydrites could have taken place not only in the gypsum-anhydrite horizon, but in the lower salt-bearing one, too.

However, it was not possible as yet to discover them in the latter, because of the complicated structure of the salt-bearing horizon.

The author made an attempt to correlate the distribution of a third group of rocks developed in the lower and middle horizons, composed of clays, with the relief of the roof of the Kungurian. When plotting all the more or less significant clay-beds on the map, all of them were found to fit within the areas of bulges of the roof of the Kungurian, while they were altogether absent in the trough-like downwarpings of the latter. Such a coincidence can by no means be merely a casual one.

As clays are in general endowed with plasticity, it should be presumed that the localization of thick clay-beds on the bulges, and their absence in the troughs, are a result of their tectonic squeezing-out from under the troughs and their shifting to bulges.

Thus, the salt tectonics of the Kungurian in the Ishimbaevo area of the Near-Ural region, expressed in a disharmony between the roof and the basement, is a result of intraformational migration not of salt alone, as it is generally assumed now, but of all the principal rocks composing the Kungurian beds: salts, anhydrites, clays.

All these rocks were squeezed-out from under the troughs, and accumulated in the bulges, evidently, according to the degree of their plasticity: salts>anhydrites, anhydrites>clays.

Like all salt tectonics, the structures of the halogen Kungurian of the Ishimbaevo area of the Near-Ural region are in general distinguished by diapirism which is, however, a peculiar, primitive and slightly developed one. Its peculiar features are: 1) the salt does not take part in the composition of the intrusions breaking through the roof, and is in general indifferent to the processes of diapirism; irrespective of whether the bulge belongs to a diapir or a non-diapir type, the salt within it does not rise above marks—285 to 300 m; 2) insignificant vertical diversions of the intrusive blocks, measuring only 200 to 250 m; 3) the absence around the piercement cores of any system of disjunctive disturbances developed to any important degree, owing to which the general anticline-like nature of the bulges is left undisturbed.

To understand the origin of the Ishimbaevo diapirs it is very important that gypsum within them attains but a very insignificant thickness (on the Smakaev block, 15 to 20 m), while the bulk of the intrusions is composed of anhydrites. It means that the hydration of anhydrites, to which frequently a decisive part in the origin of piercements is ascribed, did not play any part in this particular case.

The invasion of anhydrites into the roof is a true tectonic process, and has

been effected by the same forces which caused the squeezing-out of plastic rocks from under the troughs and their accumulation in bulges. As the upper member of the plastic rocks were anhydrites and not salt, they were intruded to the roof of the bulges; while the deeper lying salt remained unaffected by the process of intrusion.

In the course of the subsequent development of diapirism, the salt probably became also involved in the intrusions.

From all the aforesaid it follows that the usual interpretation of salt tectonics stands in need of essential corrections and further development, which becomes apparent from an analysis of the Kungurian tectonics of the Ishimbaevo area of the Near-Ural region. At the present state of affairs, this region can justly be called the classical area for the development of a general theory of the salt tectonics.

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MINERALOGY

FERGUSONITE FROM THE ADUI GROUP OF MINES IN THE URALS

By G. P. BARSANOV

(Communicated by A. E. Fersman, Member of the Academy, I. X. 1944)

In 1913 specimens of a radioactive black mineral were brought to the Mineralogical Museum of the Academy of Sciences. This mineral was found by L. I. Kryzhanovsky in mine No. 1 at the well-known occurrence of beryls and amethysts on the Adui River, and determined by him as euxenite.

In 1914 this mineral was for the first time mentioned by A. E. Fersman ([2]), when describing the paragenesis of the Adui mines, again by the name of euxenite.

Later, in 1925, he mentioned euxenite in several papers dealing with a description of the pegmatite veins of the Urals ([3,4]). Accordingly, mention of euxenite as occurring in the Adui pegmatites has been made in his cumulative review on pegmatites ([5]).

A perusal of the literature and an acquaintance with the data by Kryzhanovsky, who found it in 1913, on the ground of which this mineral had been identified as euxenite, showed that this had been done merely on account of its external appearance.

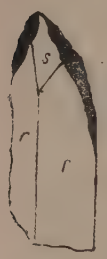
Later on, Fersman gave his assert to this diagnosis. No precise mineralogical determination of the mineral has ever been made, however. As to the specific determination of the minerals of the niobo-tantalate group according to their external characters, especially if no well-developed crystals are available, it is impracticable ([1]), because of the similarity and variability of their main physical properties.

The material under No. 21933 *etc.*, which is known in the collections of the Museum by the name of euxenite, was taken from a pegmatite, almost purely feldspar vein, cutting compact granites (Middle Urals). The body of the vein is banded-zonal, consisting of alternating zones of graphic (structural) quartz-feldspar pegmatite, and pink radially fibrous and shelly albite with voids containing beryl and other pneumatolytic minerals. A description of the vein has been given by Fersman ([4], [5]) who supposed euxenite, monazite and beryl to be concentrated chiefly at the contact between these zones. The type of pegmatite has been determined by Fersman as the IV fluor-beryllium type; paragenetically, the pegmatite is determined by the following minerals: magnetite, biotite, orthoclase-perthite, albite, quartz, muscovite, euxenite, monazite, beryl, rock crystal, amethyst.

In the specimens studied the mineral forms small irregular segregations (up to 0.25 cm^2), generally 2 to 3 mm across, grown into a pinkish and yellowish feldspar, or occasionally growing together with grey quartz of the first generation.

The impregnation of the minerals is scarce, and in the richest specimens only 3 to 5 small pairs are observed, arranged 5—3 cm apart.

Some grains are surrounded with thin reddish-brown crusts representing products of the alteration of the mineral, while the central portions usually remain fresh and unaltered. Around the grains of the mineral in the enclosing feldspar small cracks and reddish-brown halos are commonly observed; these are the effect of radioactivity. The colour of the mineral is black, sometimes with a reddish-brown hue. Lustre strong, vitreous to resinous, like that of samarskite. Fracture, bright conchoidal. Brittle. Hardness, 5.5 to 6. Specific gravity as determined by a simplified pycnometric method in alcohol from a sample of 0.25 g, is 4.96, and in browner varieties 4.88. Colour of streak light, brown-fallow. Upon the ignition of the mineral grains in a muffle furnace at 850—900° C a pyrognomic effect (self-splitting) is observed, which is evidence of the metamikt condition of the substance of the mineral. On ignition it becomes somewhat brownish, and the colour of the streak becomes brown-fallow. Optical examination by the immersion method showed that in thick splinters the mineral is not transparent, being translucent on the edges only. In thin splinters it is transparent, homogeneous, uniformly reddish-brown. In very thin grains it is light, brownish-yellow. No cleavage. Fully isotropic (metamikt). Refractive index as determined in melts was found to be above 2.06 ($n \cong 2.15$). Upon ignition and rapid cooling it passes again to the crystalline state and becomes distinctly anisotropic. In the coloration there appear brown spots. An internal granular (aggregational) structure appears. The polarization is aggregational, owing to which in some grains no complete extinction is observed. A number of grains fall into lamellar normally extinguished areas; the interference colours are grey, of the first order ($N_g - N_p \cong 0.01$). The refractive index upon ignition becomes slightly higher (about 2.2).



When examining some fine material placed in a separate test-tube, a very imperfect and broken aggregate of crystals of the mineral investigated was found intergrown with feldspar and quartz. The size of the largest crystal of the aggregate (see figure) is about 2 mm following the long axis. The drawing is made with the aid of microscope. The prismatic faces *r* make an angle of 90°.

The faces are dull, corroded, very uneven and covered with brown decomposition products. The *s* face, making a very obtuse angle with face *r*, is very distinctly observed; to measure the angle was not possible. The general habit of the crystal is a prismatic one. The aggregate is formed approximately parallel to the faces of prism *r*. The crystal undoubtedly belongs to the tetragonal (or orthorhombic?) syngony.

A chemical investigation of the composition was made by the qualitative microchemical method. Owing to the sensitivity of the reactions for individual elements, when working with solutions of equal volumes from a single initial sample (30—50 mg), some quantitative gradations may also be given: very large quantity (20—30 per cent), large amount (~10 per cent), small amount (1—5 per cent), very small amount (~1 per cent) and traces (0.01—0.001 per cent). The initial method of decomposition is fusion with KOH; Fe, Mn, Ti are determined by the routine colour reactions. The other elements are determined by a special microchemical method which consists in obtaining insoluble crystalline precipitates of compounds, each of which shows specific properties of crystals as observed under the microscope.

The results of the analysis are as follows: SiO_2, none; TiO_2, a very small amount; Al_2O_3, none; $Fe_2O_3 + FeO$, a large quantity; CaO, very small amount; MgO, none; MnO, small quantity; V_2O_3, large quantity; $Ce_2O_3 + ThO_2$, very small quantity; UO_2, small amount; $(Nb, Ta)_2O_5$, very large quantity; H_2O, small amount.

From the data obtained it follows that from the total of its physical and chemical properties the mineral can by no means be referred to euxenite, being a metamikt niobo-tantalate of yttrium and iron of tetragonal (?) syngony, into whose composition enter as minor constituents U, Mn, H_2O, and as admixtures, the group of Ce, Th and Ca. The mineral is remarkable for its almost complete

Mineral	Colour	Lustre	Hardness	Specific gravity	Syngony	Optical characters	n	$N_g - N_p$	Chemical composition		
									principal	minor	admixtures
Euxenite	Reddish-black to dark-brown	Strong, vitreous to submetallic	5—6.5, brittle	5.1—4.6, decreases upon hydration	Orthorhombic prismatic. Elongated along *C*	Transparent, in thin grains reddish-brown, isotropic (metamikt)	2.06—2.26, upon ignition 2.23	—	Nb, Ta, Ti, Y—Er, Ce—La, U	Fe, Ca Th, H_2O	Mn, Zr, Pb
Fergusonite	Brown-black to yellowish-brown	Strong, vitreous	5.5—6, brittle	5.8—4.3, decreases upon hydration	Tetragonal prismatic or bipyramidal	Transparent, reddish-brown to light brownish-yellow, isotropic (metamikt)	2.06—2.19	High, pleochroic	Nb, Ta, Y—Er	Fe, U, Ce, Th, Ca, H_2O	Mn, Ti, Zr
Fergusonite, Adui group, mine No. 1	Black to red-brown	Strong, vitreous to resinous	5.5—6, brittle	4.96—4.88	Tetragonal (?) prismatic along the *c* axis	Transparent, reddish-brown to brownish-yellow, isotropic (metamikt); anisotropic upon ignition	2.15—2.2	0.01 upon ignition	Nb, Ta, Y—Er, Fe	U, Mn, H_2O	Ce—Th, Ca, Ti

lack of titanium, which absolutely excludes the possibility to refer it to the group euxenite-polycrase, or priorite.

From the sum-total of its characters the mineral permits of no other determination than as fergusonite. Consequently, the Adui deposit is the second occurrence in the Urals, and in the USSR in general, of this rare niobo-tantalate of yttrium earths, the first occurrence of which was described by Chernik ([6]) and later on by the present author ([1]) from the Ilmen Mountains. The adjoined table, showing the comparative chemical composition and general properties of euxenite, fergusonite and of the mineral from the Adui deposit investigated by the writer, leaves no room for doubt as to the correctness of its being referred to fergusonite; the following considerations go to support this conclusion:

1) Specific gravity, hardness, colour, lustre are much similar and highly variable both in euxenite and in fergusonite, and can therefore be used as criteria of the second order only.

2) The optical properties of euxenite and fergusonite are closely similar; the higher refractive index and the characteristic crystalline structure, however, acquired by the mineral upon ignition, are features typical of fergusonite rather than euxenite.

3) The habit of the crystal is a very peculiar one (see figure). Although its imperfect development prevented it from being measured, the combination of the *r* prism with the similarly developed face *s* taken as the basal dipyramid, is highly characteristic of the crystals of fergusonite as described by Goldschmidt ([7]), and anything but characteristic of the minerals of the euxenite group.

4) Although the two minerals belong to the group of rare earths niobo-tantalates, the formula of euxenite, as suggested by Lacroix ([9]), Zambonini and Prior ([11]), can be represented as:

$$[(Nb, Ta)O_3]_2[Ca(UO_2)] \cdot [(Nb, Ta)O_3]_3(Y, Ce) \cdot [TiO_3]_2(Y, Ce)_2$$

The mineral is thus a complex niobate and titanate of rare earths, uranium, calcium, *etc.*, which can also be seen from the structural formula given by Machatschki ([10]):

$$[(Y, Ce, U, Er, Fe)(NbTi)_2O_6]$$

corresponding to the type XZ_2O_6. Indeed, in the majority of euxenite analyses ([11]) the TiO_2 content ranges from 18 to 30 per cent, *i. e.* titanium is the chief constituent element.

The investigation made by the author has shown that the mineral from the Adui contains but very small quantities of Ti, of the order of 1 per cent, its composition corresponding to that of the niobo-tantalate of yttrium earths, partly replaced by U, the Ce group, Fe, Mn, *etc.*, which is in perfect agreement with the results of analyses made by Lacroix ([9]) and Prior ([11]) and the chemical composition suggested by those writers for fergusonite.

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PHYSICAL GEOGRAPHY

BIOGEOCOENOLOGY AND PHYTOCOENOLOGY

By V. N. SUKACHEV, Member of the Academy

The modern school in geography is distinguished by that it is dealing with objects and phenomena on the surface of the Earth in their correlation and interaction. It is, therefore, the geographical landscape which is regarded as the main subject of geographical studies; and by the landscape is understood a part of territory where the elements of the lithosphere, hydrosphere, pedosphere, biosphere, and relief form over a definite area a single integer complex. Yet, aside from the fact that both the scope and contents of the term «geographical landscape» were not understood in the same way by different writers, the term was infrequently applied in an indiscriminate manner to natural phenomena having nothing in common.

In fact, if phytocoenose is considered as a part of the vegetable covering, homogeneous over a certain area, together with its animal inhabitants (zoocoenose), and with the corresponding parts of lithosphere, pedosphere, hydrosphere and atmosphere, likewise homogeneous over the same area, then it can be spoken of as an integer complex, closely held together by intrinsic correlations. For this complex I have suggested the name of geocoenose (1), or preferably biogeocoenose(2). It should be noted that by litho-pedo-hydro- and atmosphere I understand the regimes of the processes characterizing these envelopes of the Earth within a definite phytocoenose. By the regime of the atmosphere we usually understand its gaseous, thermal and light regime (the latter should preferably be defined as its radiation regime), as well as its water regime. The terms «phytocoenose» and «zoocoenose» (biocoenose, if considered as a whole) embrace all the organisms, including the microorganisms of the soil (edaphon) and of the air (aerophon) within the limits of the territory occupied by a given phytocoenose. The abovesaid can also be written in the following concise form: Biogeocoenose = Biocoenose (phytocoenose + zoocoenose) + Ecotope(1), or Biotope (2) (edaphotope + climatope)*.

In contrast to the definition of geographical landscape, the concept of relief does not enter the definition of biocoenose. The relief is essentially one of the forms of the work of the lithosphere over a given area of the Earth; it may, however, to a certain extend be correlated with the other elements of the biocoenose. Identical biogeocoenoses may, though seldom, occur under conditions of a different relief.

Biogeocoenoses are characterized by a definite system of interaction (co-action) between plants and animals, on the one hand, and between these and soil and atmosphere, on the other hand. The co-action between organisms may assume the form of struggle for life (in the broad sense, as understood by

* «Ecotope» (a term suggested by G. N. Wysotzky), or biotope (in the sense given to it by D. N. Kashkarov) embrace the regimes of definite parts of lithosphere, pedosphere and hydrosphere over a given phytocoenose.

Darwin), or in the form of favourable phenomena, mostly developed in the course of the struggle for existence. The co-action of all these agents has either physical or chemical expression; we see, therefore, no reason for the foundation of chemical biocoenology ([3]), as a separate branch of science.

The fundamental aim of the science of biogeocoenoses, or biogeocoenology, is to study all the forms of co-action referred to above, while the main purpose of biocoenology is to study the struggle for life among organisms, in all its manifestations and consequences.

Since the very moment when a naked rock becomes inhabited by the first plants and animals, begins also the formation of the biocoenose and of the soil, and consequently, of the biogeocoenose. This process, which can well be named a biogeocoenogenetic one, goes on without any interruption until there is vegetable life over the given area; its motive power consists in those intrinsic, contradictory interactions between the elements it is made of, which do never come to a standstill. And this is the process of the development of biogeocoenoses, more strictly speaking, of their self-development. According as proceeds the development of biogeocoenoses, the rôle played by the life-activity of vegetation (phytocoenose) becomes more and more important. The process of self-development of the biogeocoenose will not stop until it is destroyed by some external agent. The existence of a so-called «climax» of biogeocoenose in the sense given to it by the American and West-European writers, is therefore out of the question; as there can be no climax in the development of vegetation either ([4]). The development of phytocoenose, zoocoenose, soil and other component of the biogeocoenose is governed by specific regularities of their own.

The vegetable covering over a given area of the Earth surface can be subject to changes of three different kinds; 1) changes due to processes of self-development, the so-called syngenesis, or, more correctly, phytocoenogenesis; 2) changes connected with the process of the development of the biogeocoenose as a whole, *i. e.* due to biogeocoenogenesis; and, finally, 3) changes dependent on natural phenomena, having a casual and external influence upon the vegetation over a given area (exogenic successions) ([1]). Accordingly, the changes which may take place in a biogeocoenose, can also be subdivided into three classes: 1) development in its proper sense, *i. e.* biogeocoenogenesis; 2) changes connected with the development of a more general, broad unit, of which the given biogeocoenose is one of the components, *e. g.* the globe as a whole; 3) changes due to the development of other phenomena which may proceed in the close vicinity or be quite remote from the biogeocoenose, being in no organic relation to it; we understand here such external, foreign agents as, say, general cosmic alterations of the climate, the influence of Man, *etc.* The changes of this category can occasionally be very important and even fatal. Although the development of a biogeocoenose may assume a definite trend, or proceed with varying intensity, according to the external conditions of the medium by which it is affected, yet, the motive power of the development of a biogeocoenose is in the intrinsic contradictory co-action between its elements, rather than in the influence of the external medium. Consequently, the study of the dynamics of biogeocoenoses as a whole has to proceed along the three directions specified above; it can, of course, by no means be substituted by the study of separate elements of which the biogeocoenose is composed.

In the same way as by the term «phytocoenose» we understand a definite, concrete group of plants, the biogeocoenose has also to be conceived as an entirely concrete phenomenon which offers such a great variety under natural conditions that it is hardly possible to find two biogeocoenoses that would be identical. But the biogeocoenoses of a similar structure and following a similar course of development are united into a common type of biogeocoenose, in the same way as similar phytocoenoses are united into a common type of phytocoenose, usually called a vegetable association.

Thus, the first and fundamental classification unit in biogeocoenology is the type of the biogeocoenose, which is a term corresponding to that of vegetable

association. This unit is a very small one. As higher fundamental taxonomic units the following may be suggested: group of types of biogeocoenose, class of types of biogeocoenose, biogeocoenotic formation, group of biogeocoenotic formations, class of biogeocoenotic formations, and type of biogeocoenotic covering. When adopting lower or higher taxonomic units, it is always necessary that at their basis be laid the degree of similarity and complexity of the co-actions referred to above, and, particularly, of co-actions between the biocoenoses and the ecotype.

From the analysis of the nature of the biogeocoenose it must be clear that it is necessary to discriminate between the biogeocoenose and the geographical landscape, and that, accordingly, biogeocoenology must not be confounded with the science of landscape. Aside from the fact that the concept of the landscape embraces the form of the relief, and that even the narrowest, elementary landscape in the interpretation of Polynov ([5]), Pervukhin ([6]) and Ponomarev([7]), or geographical landscapes of the 2nd order in the sense given to them by Berg ([8]), are considerably wider than the concept of biogeocoenose, the geographical landscape and the biogeocoenoses are two conceptions different in principle. In the science of landscape or, if we are to adopt the term which has recently been suggested by Berg, the «science of geographical aspects», the taxonomic units coincide with the units which are adopted in the geographical subdivision. Unlike this, the units of biogeocoenology do not coincide with them. Thus, the geographical landscapes of the 2nd order are united by Berg into geographical landscapes of the 1st order; the latter are further on united into landscape zones, such as the tayga, the tundra, the forest-steppe, the steppe, and so on. When working out a generalized elementary landscape of West-Siberian birch-grove, Ponomarev likewise unites those groves with the groves of other regions in a single geographical landscape, that of the forests of forest-steppe depressions. However, not only the forest-steppe, where areas of forest and of steppe, *i. e.* of two quite remote biogeocoenoses, are found to co-exist side by side, but also the West-Siberian groves do often display small swampy areas surrounded with forest; in a single biogeocoenose we observe here two elements, differing sharply as to their structure and character. The very concept of forest-steppe is a concept borrowed from the science of landscape, rather than a biogeocoenotic term.

Such concepts as meadow, steppe, forest, swamp, *etc.*, are biogeocoenotic and not phytocoenotic concepts: they embrace meadow-, steppe-, woody, swampy, *etc.* vegetations, which are composed of phytocoenoses. The application of the term of «forest» or «steppe» to an entire zone also implies a lack of precision; it is more correct to speak of a forest- or steppe-zone as landscape conceptions which may comprise other types of biogeocoenotic covering, though the predominant type will be the steppe or the forest, respectively. The concept of biogeocoenose approaches that of ecosystem, suggested by Tansley ([9]).

We thus see that the classification problem in biogeocoenology and in the science of landscape is based upon entirely different foundations.

From what has been said above it may clearly be seen that although the phytocoenose enters into the biogeocoenose as one of its components, phytocoenology is not a part of biogeocoenology, for the life of the phytocoenose and that of the biogeocoenose obeys to different regularities. But biogeocoenology and the science of landscape are also two different disciplines, which make use of different working methods.

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GENETICS

TETRAPLOID KOK-SAGHYZ AS A VARIETY OF IMPROVED PRODUCTIVITY

By M. S. NAVASHIN, E. N. GERASSIMOVA and A. F. CHEREDNICHENKO

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The present paper contains a brief account of our work on the tetraploid kok-saghyz, carried on during the last three years. The main object of the work was by applying tetraploidy to raise the productivity of this most important rubber-plant. In the course of the work we were assisted by Miss G. M. Belajeva.

The first observations made on the tetraploid kok-saghyz immediately upon its experimental production (1) have shown that there existed good prospects for raising productivity through tetraploidy. The considerable amount of data which have been accumulated since permit now of definite conclusions.

The tetraploids of kok-saghyz differ morphologically from the diploids in a number of general characters typical of the majority of tetraploids. These distinguishing characteristics originate from an increase of all the cells, organs and of the plant as a whole, which results from the doubling of the chromosome complement. Besides, there is a strong increase in the size of the achenes which develop on the tetraploid plants. The increase of all the dimensions is so conspicuous that, in connexion with some other peculiar characters (greater relative width of the leaves, which are more fleshly and of darker green, *etc.*), makes it possible at once to discriminate between tetraploids and diploids.

The tetraploid kok-saghyz thus represents a very distinct form which remains as such in spite of the great individual variation peculiar to this species.

It was in 1941 that we were able to study *en gros* the tetraploid kok-saghyz produced by us; at that time we had already developed, at the Kropotovo Biological Station of the Institute of Cytology of the Academy of Sciences of the USSR, a plantation consisting of over 20 000 of tetraploid plants. In the autumn of 1941 the work was transferred to Kazakhstan. By that time it has become obvious that the tetraploid kok-saghyz possessed a number of economically valuable characteristics, the principal of which are greatly increased roots, an increased diameter of the laticiferous vessels, enlarged seeds and robust seedlings. Thanks to the support of the Trust of Plant-rubber, the work could be considerably extended in 1943. At the present time the tetraploid kok-saghyz is being propagated at the seed-growing state farms of the Trust (No. 12 in Southern Kazakhstan and the Petrinsky farm in the Kursk region).

A c h e n e s a n d S e e d l i n g s. The weight of 1000 achenes of the tetraploid kok-saghyz varies from 0.6 g to 1.0 g and more, *i. e.* exceeds 1.5—2 times the absolute weight of the diploid. This is a decisive advantage if the well-known sensitivity of kok-saghyz to the depth of sowing be taken into account. The high individual variation in the weight of seeds makes one expect that this valuable

property may be developed further by selection. The increase in weight is due, however, in a somewhat greater measure to the development of the covers of the achene rather than to a greater weight of the embryo. Between the loose pericarp and the seed in the tetraploid achene there is a small cavity which is filled with water when the achene swells. The total amount of water absorbed attains, therefore, 180 per cent (to the weight of the achene) as contrasted with 120 per cent in the diploid kok-saghyz.

The germinating power and the germinating force in the normally developed tetraploid achenes are not inferior to those in the diploid. Under favourable conditions sowing gives 80 per cent of seedlings and even more. The seedlings are strikingly robust and strong; owing to the latter property, they can lift the soil which covers them. As regards the tempo of the development of the seedlings, no differences could be observed. There were no peculiarities in the reaction of the tetraploid seeds to stratification or soaking in water before sowing either.

Development of the Plants. In the test-sowings made at the Botanical Gardens at Alma-Ata (Kazakhstan) and at the No. 12 State Farm (Southern Kazakhstan) no differences in the rate of development of the tetraploid and diploid plant were noticed. All the developmental stages ensued nearly at the same time in both. Neither could there be noticed any difference

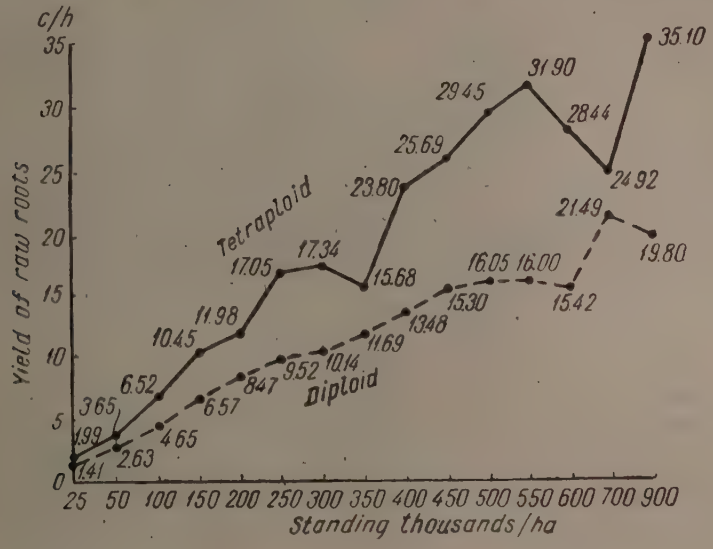

Fig. 1

in the proportion of plants blooming in the first season or in the dates of entering into summer rest. Thus, the tetraploid kok-saghyz does not show any deviation in the rhythm of its development, which is in contrast with the majority of other tetraploids known. The possibility is not excluded, however, that such slight differences as may exist are subdued by the strong individual variation.

Yield of the Roots and Average Weight of a Root. More extensive data were obtained from comparative sowings made in 1943 at the Rubber State Farm No. 12. As the soil was highly variable (saline spots), the yield was recorded for each metre separately. Fig. 1 presents these comparative data (reference being made to the standard material grown from seed produced at the No. 12 State Farm).

Fig. 2 gives an idea of the average weight of an individual root from the comparative sowing.

From the data presented above it follows that the tetraploid invariably surpasses the diploid both in the yield of roots and in the weight of an individual

root. This difference is very significant and attains on the average 60 per cent.

Rubber Content. The data obtained by us have shown that the roots of the tetraploid kok-saghyz do not differ from those of the diploid in rubber content, the latter being during the first year of life from 5 to 8 per cent (to the absolutely dry weight). As may be seen from the following table, the difference, if any, is rather in favour of the tetraploid. As shown by the analyses

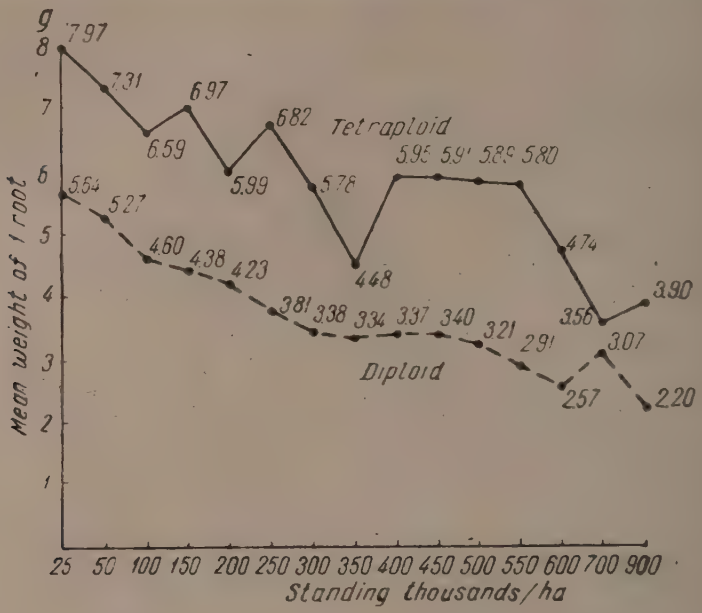

Fig. 2

which we owe to the scientific worker of the Institute for Rubber Investigation, Obolenskaya, the tetraploid very closely approaches in rubber content the diploid (the average values being 1.76 and 1.66 per cent, respectively). The low absolute figures should be explained by the growing conditions and by the immaturity of the plants.

Number of recorded metres of sowing													
	Tetraploid	1	8	8	8	5	9	6	6	9	5	7	5
	Diploid	7	8	7	9	7	9	9	7	10	8	9	6
Rubber % in roots	Tetraploid	0.43	1.13	1.24	1.40	1.47	1.57	1.59	1.98	1.99	1.42	2.55	3.34
	Diploid	0.62	1.18	1.33	1.44	1.41	1.51	1.59	1.66	1.94	2.15	2.26	2.88

The original suggestion as to the possibility of a reduced rubber content in the tetraploid (²) has thus not been supported by direct observations.

As shown by the work of Nichiporovich (²) and our own studies, the roots of the tetraploid kok-saghyz are distinguished by having wider laticiferous vessels. This results in a more rapid flow of the latex, which should be considered a technical advantage; when the tetraploid roots are sectioned, the latex flows out far more rapidly and abundantly than from the diploid ones. A special study has shown that the concentration of the latex in a tetraploid does not differ from that in a diploid.

Prospects of Exploiting Tetraploid Kok-saghyz for Commercial Growing. The following characters are beyond doubt to the advantage of the tetraploid: increased seed size; more robust seedlings and larger roots, their elements included; increase in the mass of the roots, which facilitates their lifting and treatment; increase in the diameter of the laticiferous vessels and an easier gain of the latex, and an increase in the diameter of the rubber filaments upon coagulation. To these advantages should be added a steeper conical shape of the root and a higher grade of polymerization of rubber (according to L. G. Dobrunov). Besides, as shown by our rather extensive observations, the amplitude of the individual variation in a number of characters is noticeably greater in the tetraploid kok-saghyz than in the diploid. This permits to look forward to a greater effect of selection.

The tetraploid kok-saghyz should thus be considered a valuable material which promises good results in the breeding for better varieties of this most important crop. However, it should not be forgotten that the tetraploid still has certain undesirable characteristics. The principal defect which is connected with a higher variability, consists in a reduced fertility of a certain fraction of plants. As shown by special studies ([3]), this defect can surely be removed by selection. Besides its harm is no very great one, since even to-day the tetraploid has an entirely satisfactory coefficient of reproduction; thus, the yield of seeds under favourable surrounding conditions attains on the first year 0.2 g per plant, which, with 400 000 plants per hectare, would yield 80 kg of seed per hectare. Another defect, although an insignificant one, is the higher water content of the roots (26.2 per cent of dry weight *vs.* 28.9 per cent).

It can safely be predicted, however, that correct breeding work will not only eliminate all these defects, but will produce from the tetraploid kok-saghyz a variety endowed with qualities inattainable by the diploid; it should be remembered that natural selection had created thousands of polyploid species possessing decisive advantage over their diploid predecessors. Even in its present state, as a population showing a sharp shift toward improvement, it will beyond doubt secure a considerable additional yield. As noted above, the tetraploid material is now being propagated and has been subjected to large-scale variety tests. The results of geographical sowings are of great interest, since they may reveal a specific answer of the tetraploid to the surroundings.

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PLANT PHYSIOLOGY

INFLUENCE OF COPPER UPON THE POTATO PLANT

By K. SUKHORUKOV and E. KLING

(Communicated by A. A. Richter, Member of the Academy, 3. VIII. 1944)

Copper is accumulated in the tubers of the potato plant in quantities amounting to $4 \cdot 10^{-4}$(1). According to data by Kubovitsch and Warburg's interpretation, this element enters there into the composition of the cupro-proteids which are operative in the respiration process. As compared with the other mineral elements, the amount of copper contained in the potato plant is insignificant. This may be taken to mean that the potato plant receives under field conditions a sufficient amount of copper, so that there is no shortage with respect to this element (7).

However, our observations carried on during several years have shown the potato plant to respond in a rather distinct manner to additional copper diet even if grown under field conditions.

Formation of Tuber as Affected by Copper. For the experiment were selected identical fragments of tubers, *viz.*, «tops» of the variety Berlichingen. The fragments were kept for some time under room conditions in the light and showed noticeable withering; however, they sprouted normally. After having been rinsed in water, the fragments were transferred to containers with a 0.001 *M* (0.025 per cent) solution of copper sulphate, and remained there during ten hours. Then they were slightly rinsed in water and planted into moistened soil contained in boxes placed in front of a window opening to the South. Along with the experiments, water- and dry controls were arranged. Planting was done on the 1st of May, 1943. Within fifteen days noticeable differences appeared. Those plants which have received copper, developed broader leaf-blades of a deeper green; the growth in them was delayed. On the other hand, the two controls developed long shoots bearing leaves of a paler green. On the 23rd of May, after warm weather set in, the plants were transferred to the experiment lot of the Botanical Gardens of the Tomsk State University. The differences noted above persisted till the 25th of August, when the harvest took place. The yield of tubers was recorded for 10 similar hills (Table 1).

Table 1

Yield of Tubers (Field experiment involving 10 plants)

	Number of tubers				Weight			
	50 g and more	50—30 g	less than 30 g	total	50 g and more	50—30 g	less than 30 g	total
Plants which received copper .	24	24	56	104	2.28	0.80	0.66	3.74
Water control . .	16	60	104	180	1.44	2.20	0.96	4.60
Dry control . . .	12	24	160	196	1.56	0.96	0.96	3.48

It may be seen from the above figures that copper does not increase the total yield of tubers; it accelerates, however, the formation of tubers and enhances the proportion of large tubers. The following laboratory experiment was arranged to check this. Sprouting tubers weighing 30 g each were treated during 24 hours with a 0.001 *M* solution of copper sulphate; the control was kept for the same period in water. After treatment the tubers were transferred to moistened containers and placed into a dark room at a temperature of 18°C. After 40 days the shoots were measured, and the newly formed tubers recorded (Table 2).

Table 2

Effect of Copper upon Growth and Formation of Tubers (Laboratory experiment)

	Average length of shoot, cm	Total number of tubers	Total weight of tubers, g	Average weight of tuber, g
Plants which received copper	20.3	40	31.9	0.80
Control	51.2	9	4.0	0.44

The results of the laboratory experiment thus went to confirm the field experiment, *viz.* that the effect of copper is to detain growth and enhance formation of tubers.

Effect of Copper upon Resistance to *Phytophthora infestans*. The plants which have received copper and the controls (variety Berlichingen) were tested for resistance to *Phytophthora*. For inoculation were used infected leaves brought from potato fields in the vicinity of Tomsk. Under all possible precautions, the plants were inoculated with a suspension of the conidia of a highly virulent fungus. Ten leaves were inoculated each time; the records were taken four days after inoculation. The results are summarized in Table 3.

Table 3

Development of *Ph. infestans* upon Inoculated Leaves

	Necrotic area of leaf, in %	Kind of necrosis	Development of *Ph. infestans*
Plants which received copper	30	Sharply delimited dark necroses	Poor growth of micelium, slight formation of conidia
Control	100	Darkening of the whole leaf	Good growth of micelium, normal formation of conidia

As seen from Table 3, copper has noticeably augmented the resistance of the plants to infection, and judging by the characters of the injury, they had acquired the properties of immune plants. There exist two different opinions with regard to the effect of copper upon the immunity of plants. The one, widely spread opinion is that copper does not «make» the plant immune, but has a direct toxic effect upon the parasite (Laurent)(6). According to the other opinion, in certain quantities copper has a beneficial effect upon the general condition of the plant and, in addition, augments its resistance to disease(9). Minaeva studied in 1939—40, under the guidance of Sukhorukov, the reaction of *Phytophthora infestans* to the various concentrations of copper in nutritive media. *Phytophthora* was sown on oats-agar with addition of copper sulphate at concentrations from 0 to 2.5 per cent. It was borne out with sufficient clarity that the growth of *Phytophthora* was stimulated in the presence of copper at a concentration of 0.125 per cent ($\frac{1}{200}$ *M*), and depressed by stronger concentrations. We are inlined,

therefore, to explain the increase in the resistance which was noted by us, by a change in the condition of the organism under the influence of copper. Our hypothesis is supported by the results of the determination of peroxidase in the leaves with the aid of a method, described elsewhere (4). The activity is expressed in cm^3 of 0.1 *N* $KMnO_4$ per 1 g of fresh leaves.

Plants which received copper gave 18.1; controls, 12.2.

The activity of the peroxidase points to an increase in oxidation potential in the experimental plants. According to Grechushnikov (2), the cultivated varieties and the wild species of potato which are immune to *Phytophthora infestans* show a higher activity of peroxidase. There are grounds, therefore, to believe that copper in this case does not protect the plant directly by its toxic action, but rather indirectly, through a change in the physiological properties of the plant. As a matter of course, the above-said should not be taken to mean that those protective measures where copper is acting directly upon the parasite, are to be questioned. Some of the potato growers at Tomsk practised at our advice addition of copper to soils under potato. The overground parts of the potato plants were periodically sprinkled with a 0.02 per cent solution of copper sulphate. The results were favourable: the plants have shown better resistance to the fungus and yielded a greater number of large tubers.

An additional introduction of copper into the potato being a useful measure, deserves a practical study, especially for those regions where «*Phytophthora* seasons» are of frequent reoccurrence.

There seems to be a possibility of producing a shift in the redox properties of the plants by introducing certain elements into them. Richter and Sukhorukov (3), working with sunflower, succeeded in augmenting the activity of the peroxidases and in reducing immunity to *Orobanche cumana* by introducing iodine. Our efforts to increase the oxidation potential of the cells by stimulating potato with iodine (the potato plant is accumulating iodine in its normal condition (13)) revealed a peculiar reaction of the plant to the introduction of this element. Those plants which had received iodine, showed noticeably accelerated growth, and their leaves assumed a paler tint, *i. e.* the external reaction was opposite to that caused by copper. This somewhat unexpected phenomenon received its explanation in the work by Levis(8) on the effect of copper upon *Azotobacter*. This writer has observed a reduction in the effect of copper in the presence of iodine, which he explains by the chemical interaction between the two elements within the organism of the plant. If it is so, it seems possible to inhibit the physiologically active substances by affecting the organism in a manner analogous to that employed by us.

It has been thus demonstrated that an additional quantity of copper introduced into the plant detains the growth and stimulates chlorophyll formation. Probably, this reaction to copper is typical of many higher plants (see monographs [5], [6], [10], [11]). The specific reaction of the potato plant is manifest in an accelerated formation of tubers and in an increased resistance to *Phytophthora infestans*.

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PLANT PHYSIOLOGY

EFFECT OF DAY-LENGTH UPON THE FORMATION OF ROOT NODULES ON THE ROOTS OF LEGUMINOUS PLANTS

By M. CH. ČAJLACHJAN and A. A. MEGRABJAN

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The length of diurnal illumination has a strong influence upon the plants, its variation causing changes in the rate of blooming and fruiting, in the intensity of growth processes and in anatomical and morphological characters.

It was quite natural, therefore, to suppose that the susceptibility of leguminous plants to the infection by root-bacteria could be influenced by controlling the length of the day.

The first attempt in this direction was made by Eaton (6) who grew soy-beans and peas at a day-length varying from three hours to continuous illumination; he observed the formation of root nodules upon the roots of all the experimental plants, and was unable, therefore, to draw any definite conclusion. Moshkov (2) has found subsequently that root nodules were formed upon the roots of *Rubinia pseudacacia* only in the case where the length of the day did not exceed twelve hours, while in another leguminous plant, *Halimodendron argenteum* Fish., the nodules were formed, on the contrary, with a short, twelve-hour day only, and did not appear under the conditions of a normal long day. In this instance, no correlation between the development of root nodules and the length of the day could be established.

The existence of such a regular relation between the influence of the length of the day and the formation of the root nodules might be anticipated, however, on the ground of the physiological changes caused by the variation in the length of the day. It has been shown by Čajlachjan (4) that the growth and the accumulation of dry matter is independent of the photoperiodic reaction and proceeds with the higher intensity, the longer is the day. The physiological interpretation of this regularity is to be found in the data on the accumulation of the carbohydrates and the formation of the growth substances, as connected with photoperiodism. The analyses by Knodel (7), Čhajlachjan (4), Murneek (8) and others have shown that the content of carbohydrates is always higher in the plants of the long day as compared with those of the short day. On the other hand, studies by Čajlachjan and Ždanova (4,5,1) have discovered the fact that the production of the growth hormones was also considerably higher under the long day in all the plants including the *Leguminosae*.

In so far as the principal condition of the symbiotic relations between the leguminous plants and the nodule bacteria consists in the rich carbohydrate content, it might be supposed that a more intense formation of nodules should take place on the roots of the plants grown under long-day than under short-day conditions.

To check up this hypothesis we carried on experiments in the greenhouse of the Armenian Agricultural Institute in 1944.

The following plants were chosen as a material for the experiments: beans (*Phaseolus vulgaris*) and soy-beans (*Soja hyspida*), both being plants of a short day; *Ervum vicia*, vetch (*Vicia sativa*) and *Phaseolus aureus*, all of them being plants of long day. The seeds of the above species were planted on the 24th of May in glass containers of 3—4 kg capacity, filled with washed quartz sand, fertilized with the Prianishnikov mixture containing half the normal ration of nitrogen. Each of the species was planted in sixteen containers; eight of them were left under the conditions of natural long day at Erevan; the other eight were kept under the conditions of an artificial 10-hour-long day. The short day was brought about with the aid of plywood light-proof boxes by which the plants were covered from 6 o'clock in the evening till 8 o'clock in the next morning; the curtailing of the day was started on the 6th of June, immediately after the completion of germination and thinning of the seedlings. In the containers with the beans, *Ervum* and *Phaseolus aureus* there were left five plants in each; in the case of vetch, four plants, and only three plants of soy-bean were left per container. Prior to planting, the seeds were inoculated with the corresponding culture of the root nodule bacteria for half an hour.

In the course of the experiments the plants were regularly watered, the water content of the sand being brought to 70 per cent of the full water capacity; systematic measurements of the growth and phenological observations were also regularly made. The influence of the duration of the day upon the condition of the plants could very soon be noticed; in all the species the plants grown at a long day were taller and more robust than those grown under short-day conditions. The photoperiodic reaction differed as to the species: beans and soy flowered and set fruit at a short day, while under the conditions of long day beans remained at the stage of vegetative growth and the soy was considerably behind in butonization and flowering. *Ervum*, vetch and *Phaseolus aureus* entered flowering and fruit-setting with the long day: *Ervum* formed no flower buds and did not bloom with the short day. Data on the effect of day-length are to be found in Table 1.

The development of nodules upon the roots of the plants was recorded at the following three dates: 1) 15 days after the beginning of the treatment with a short day; 2) at the time of budding of the plants which were at the photoperiod optimal for their development; 3) at the flowering stage, and 4) at the beginning of the maturation stage. Each test comprised four containers, two of them kept at a long day, and two—at a short day. The data on the dry weight of the plants (gms), on the number and weight (gms) of the root nodules per plant are presented in Table 2.

Table 2 shows in an explicit form the main results of the experiments, *viz.* that the formation of the root nodules, as judged by their total weight, is more intense with the long day in all the plant species tested and at various developmental stages. This intensified development of the root nodules agrees well with the intenser growth of the plant and with the accumulation of dry matter under the conditions of the long day: it does not depend upon the kind of the photoperiodic reaction of the plant, *i. e.* upon whether it blooms sooner with a short day or a long day. In all the plants grown at a long day the root nodules were larger than in those grown under short-day conditions.

It should also be observed that there is a relation between the intensity of growth, the accumulation of dry matter, on the one hand, and the formation of root nodules, on the other hand, not only within a single species under varying length of the day, but in different species grown under identical conditions, too. This may clearly be seen on the diagram.

The internal agent which creates favourable conditions for the infection with the root-nodule bacteria is undoubtedly the high content of carbohydrate in plants grown at a long day. Another internal factor favouring these symbiotic relations is probably the high content of the growth hormones formed in the plants under long-day conditions.

Table 1

Effect of Day-length upon Growth and Development of Leguminous Plants

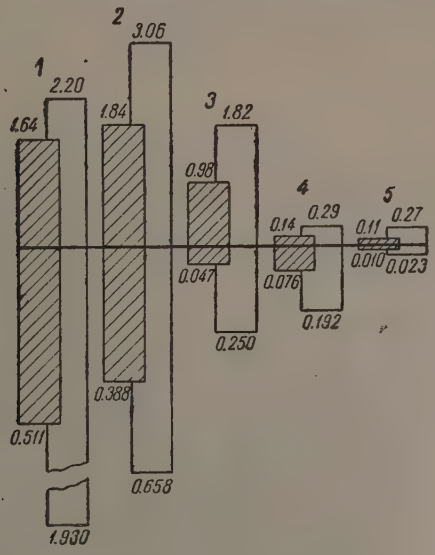
Plant	Day-length	Number of plants recorded	Height of plants towards the close of exper., cm	Budding		Flowering		Number of pods per plant	Developmental stage at the moment of harvest
				beginning	50%	beginning	50%		
Plants of short day									
Beans	L	10	69	none	none	none	none	none	Vegetative growth
	S	10	24	24.VI	27.VI	3.VII	3.VII	2.6	Beginning of maturation
Soy-beans	L	6	32	26.VII	26.VII	30.VII	30.VII	none	Flowering
	S	6	33	16.VII	16.VII	20.VII	20.VII	2.9	Beginning of maturation
Plants of long day									
Ervum Victa	L	10	24	24.VI	26.VI	29.VI	1.VII	1.2	Beginning of maturation
	S	10	15	none	none	none	none	none	Vegetative growth
Vetch	L	8	28	22.VII	22.VII	27.VII	27.VII	0.2	Beginning of maturation
	S	8	23	26.VII	26.VII	1.VIII	1.VIII	none	Flowering
Phaseolus aureus	L	10	20	15.VII	15.VII	22.VII	22.VII	2.2	Beginning of maturation
	S	10	18	18.VII	18.VII	24.VII	24.VII	2.6	» » »

Table 2

Effect of Day-length upon Accumulation of Dry Matter and Development of Root Nodules in Leguminous Plants

Plant	Day-length	At the stage of vegetative growth			At the phase of budding			At the flowering phase			At the phase of maturation		
		Weight of individual plant, g	Root nodules		Weight of individual plant, g	Root nodules		Weight of individual plant, g	Root nodules		Weight of individual plant, g	Root nodules	
			number	weight, g		number	weight, g		number	weight, g		number	weight, g
Plants of short day													
Beans	L	1.00	58	0.155	1.33	131	0.529	1.35	193	0.840	2.20	281	1.930
	S	0.81	17	0.016	0.98	58	0.135	1.16	61	0.245	1.64	110	0.511
Soy-beans	L	0.20	none	none	0.63	1	0.006	1.26	23	0.054	1.82	59	0.250
	S	0.21	»	»	0.32	2	0.006	0.88	8	0.010	0.98	15	0.047
Plants of long day													
Ervum Vicia	L	0.11	14	0.014	0.16	12	0.014	0.20	17	0.017	0.27	25	0.023
	S	0.07	9	0.008	0.12	12	0.011	0.09	6	0.007	0.11	8	0.010
Vetch	L	0.11	31	0.048	0.11	11	0.023	0.11	18	0.034	0.29	90	0.192
	S	0.06	8	0.012	0.09	15	0.011	0.09	21	0.023	0.14	46	0.076
Phaseolus aureus	L	0.45	4	0.010	2.10	42	0.471	2.42	46	0.628	3.06	51	0.658
	S	0.28	4	0.010	1.25	38	0.344	1.83	28	0.365	1.84	35	0.388

The formation of root nodules is notably preceded by the growth of the cortical parenchyme and the division of the cells of pericycle, which requires large amounts of growth hormones. Direct experiments by Thimann (9) have shown that the formation of root nodules on the roots of leguminous plants is connected with the intensity of the production of the growth hormones, since these are excreted in large quantities by the root nodules cut off the plant. Although Thimann



Accumulation of dry matter and formation of root nodules in leguminous plants at a long day (unshaded) and at a short day (shaded). *1*, beans; *2*, *Phaseolus aureus*; *3*, soy; *4*, vetch: *5*, *Ervum Vicia*. Above, dry weight of an individual plant (gms); below, total weight of root nodules per plant (gms)

himself and Cholodny following him (3) assume that these growth hormones are the products of bacteria, there is reason to believe that the leguminous plants themselves are not inactive in this respect, the high content of growth hormones in their root parenchyme being the second condition favouring the formation of root nodules.

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ENDOCRINOLOGY

FUNCTION OF PARATHYROID GLANDS IN *AMPHIBIA*

By A. N. STUDITSKY

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The function of the parathyroid glands or the epithelial bodies in *Amphibia* is far from having been thoroughly studied. Experiments involving the removal of these glands in different *Urodela* and *Anura* species gave quite contradictory results. On the one hand, the experiments on parathyroidectomy are known to have demonstrated the lack of any effect of the operation in *Rana temporaria*, *Salamandra atra*, *S. maculosa* and *Triton alpestris*. On the other hand, similar and quite reliable experiments on the removal of parathyroid glands in *Rana cantesbiana* [1] have brought about the characteristic tetanus in the experimental animals which perished some hours after operation.

To answer the question whether the parathyroid glands in *Amphibia* are playing a rôle of vital importance, the experiments on the removal of these glands were carried out in *Triton stellatus* from *Urodela*, and *Rana ridibunda* from *Anura*. These species were chosen because of their large body size.

The technique adopted was the surgical one. The removal of all four epithelial bodies was performed under ether narcosis, through two abdominal incisions, in the right and the left side of the body. The epithelial bodies in the frog can easily be found, owing to their location close to the jugular bodies. The latter, in turn, can easily be found because of their characteristic appearance, location and large size. It is rather difficult to find the epithelial bodies in *Triton stellatus*, and an operation involving their removal, owing to their small size, offers certain difficulties that cannot be overcome without persistent training.

The operations were carried out under a binocular microscope. Each gland removed was studied histologically. After some preliminary training, the epithelial bodies both in frog and triton can easily be recognized, and distinguished from any other organ in this region of the body.

The species used in the experiments are characterized by a superficial arrangement of blood vessels in the glands in question. They form a peculiar capillary network which may be seen fairly well on the surface of the gland when freshly dissected. Tritons and frogs have, as a rule, only two pairs of epithelial bodies. In about 20 tritons and 100 frogs operated by us, the number of epithelial bodies found was invariable two. No accessory glands were found in the material under consideration.

Operations on frogs were performed since November, 1940, up to May, 1941. The behaviour of the frogs operated and recovered from narcosis did not differ from that of the normal ones. During a whole winter period (since November to May) the operated frogs were kept in the 6-liter broad-spout bottles, without any food. A single group of them was occasionally fed some meat. No response to the operation was observed in the behaviour of the animals of this group.

The operated frogs were transferred to a terrarium with a reservoir where they

were allowed to stay until winter 1941. Normal egg deposition took place in May, 1941.

No disturbance in bone formation or calcification of the skeleton was observed in the operated frogs. To make some observations on the regenerative bone-formations the *tibiale*, *fibulare* and *tibia fibula* were broken in a part of the operated frogs. The rate of knitting of the bones in the animals of this group did not differ from that in the controls. The stages of formation of the bone calluses were the typical ones, including the formation of a great chondroid adhesion, its gradual replacement by the bone tissue and, at last, an intensive calcifica-

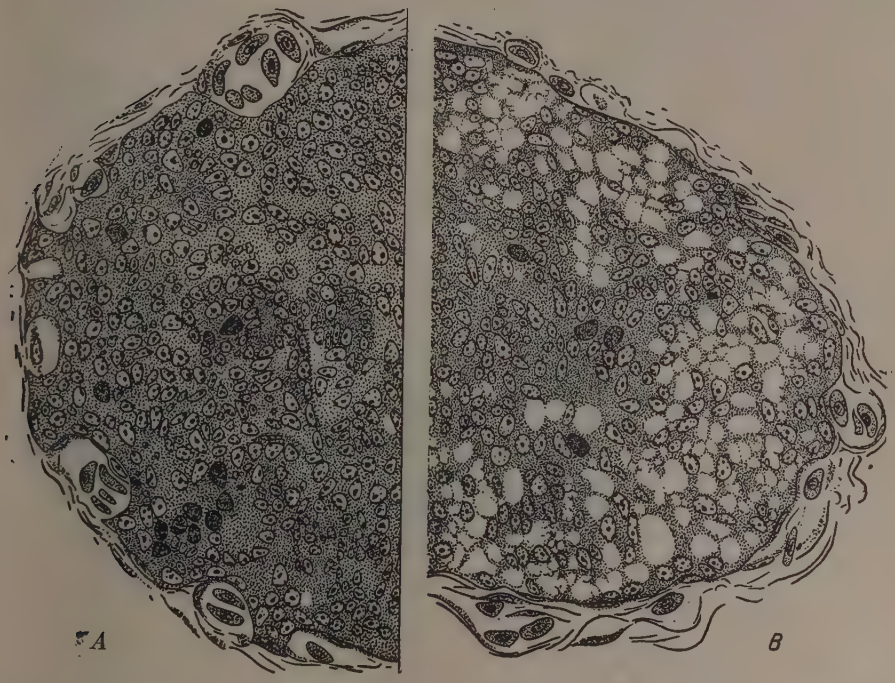

Fig. 1. Parathyroid gland in *Rana temporaria*. *A*, in winter: *B*, in summer. Excitation is manifested as a protoplasm vacuolization

tion. The whole period of formation of fairly calcified bone callus did not exceed four months. No morphological changes in the skeleton of the operated frogs were observed either.

The inner organs did not show any changes. The liver and kidneys were normal both in colour and density. The pathological changes in the eye-structure observed in mammals after parathyroidectomy were not observed in the operated frogs throughout nearly a year of observation.

Observations on the parathyroidectomized tritons led to similar conclusions. During the period of observation no difference was found in the structure of the inner organs and eyes, nor in the animals' behaviour.

On the basis of the results obtained it is possible to conclude that the epithelial bodies do not play any important rôle in the physiology of the animals of the species studied. However, it would be erroneous to regard them being destitute of such functions as make these organs of vital importance for mammals. It is known that the principal function of the parathyroid glands in mammals is to keep the calcium metabolism of the organism at an optimal level. Parathyroidectomy of the epithelial bodies in mammals leads to a fatal decrease in calcium content, causing tetanus and often the death of the animals.

The ancestors of the amphibian, the fish, are destitute of the parathyroid glands. It is possible to suggest that the regulation of calcium metabolism in this most ancient group of vertebrates is brought about with the help of an exogeneous factor, the vitamin D, that is accumulated in enormous amounts, especially in *Teleostei* (2). As they became terrestrial animals and the specific gravity of their body increased accordingly, there was developed a robust skeleton rich in calcium salts. The calcification of the skeleton in *Amphibia* is possibly still brought about under the influence of vitamin D. However, alongside of this mechanism, the parathyroid hormone is also coming into operation. At a definite period of life it is controlling the calcium content in the internal environments of the organism, mobilizing the skeleton calcium, when necessary.

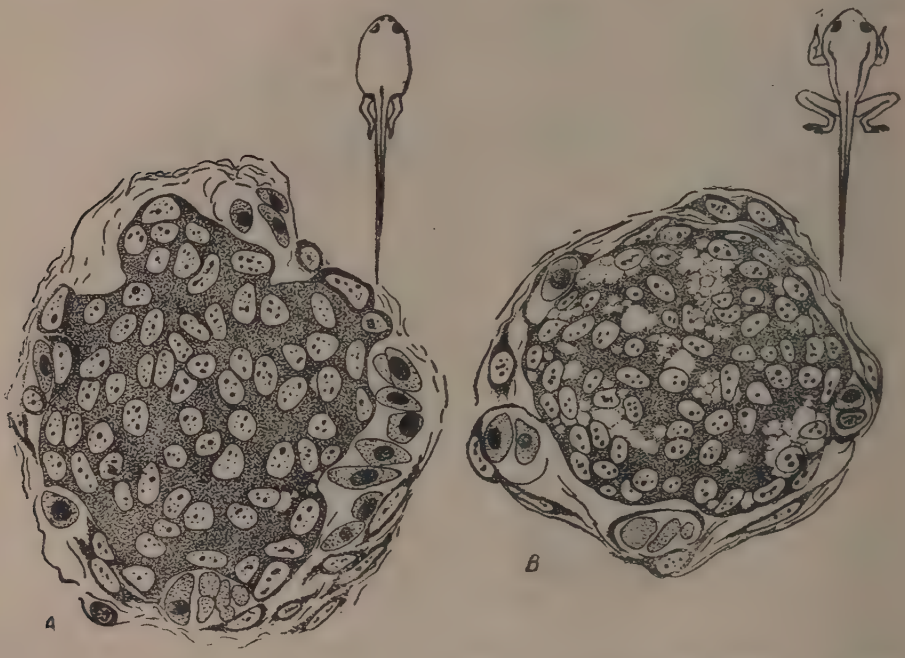

Fig. 2. Parathyroid gland of *Rana temporaria* during metamorphosis. Excitation is manifested in a protoplasm vacuolization during the calcification of skeleton (*B*)

The investigation of the seasonal and ontogenetic changes in the structure of the frog epithelial bodies has revealed certain signs of excitation—protoplasm liquefaction, at those periods which may be considered as the critical phases of calcium metabolism. Waggener has shown that the maximum protoplasm liquefaction, cytolysis, and cell degeneration in the epithelial bodies in *Rana cantesbiana* takes place during the spring months and is completed by the end of May (3).

On the basis of histological examination I have found similar liquefaction processes of the protoplasm, pointing to the excitation of the parathyroid glands, in *Rana temporaria* and *Rana ridibunda* during the spring and summer periods (Fig. 1). According to Kovalsky (4), calcium concentration during the winter and autumn periods is equal to 5.25—7.45 mg per cent, reaching the maximum in the spring (8.41 mg per cent), and decreasing to a certain degree in the summer (up to 8.09 mg per cent). There is every reason to connect the variation in calcium content with the changes in the structure of the epithelial bodies, as a sign of their excitation.

On the other hand, I have found that the characteristic liquefaction of protoplasm and the beginning of the skeletal calcification is also observed in the epithelial bodies of *Rana temporaria* and *Rana ridibunda* during the ontogenetic metamorphosis (Fig. 2). These findings also point to the participation of the epithelial bodies in the calcium metabolism.

The data here discussed seem to show that the epithelial bodies in the amphibian species studied are playing a definite part in the regulation of calcium metabolism, although these organs by themselves are not necessary for the functioning of the organism. At the same time, their function did not yet reach the importance which in this group of vertebrates evidently belongs to another regulator of calcium metabolism, *viz.* vitamin D.

The evolution of calcium metabolism in vertebrates is evidently going on in the direction of an increasing rôle of endogeneous mechanism in the inner environments of the organism; this mechanism is represented by the hormone of the parathyroid gland.

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PHYSIOLOGY

BIOELECTRIC PHENOMENA AS VIEWED IN RELATION TO CHEMICAL DYNAMICS OF THE CELL

By CH. S. KOSHTOJANZ, Member of the Academy of Sciences of the Armenian SSR

In our earlier papers it has already been pointed out ([1–4]) that a block of the transmission of nervous excitation, provoked by different inhibitors of the carbohydrate metabolism, was in many cases observed to coincide with a block of the chemodynamics of the cell at a definite stage of the latter. The effect of these substances when applied to the cardiac or sceletal muscle of the frog is to disturb the normal course of the transfer of excitation from the corresponding nerve to the tissues. By properly chosing both the concentration of the solution and the duration of treatment, we succeeded in blocking the transmission of nervous excitation; this seemed to be due to the inhibition by the paralysants of definite links in the chemodynamics of the cell, which are necessary for the transition from the «chemodynamics of rest» to the «chemodynamics of excitation».

We have earlier described the «fluoride block of excitation» which coincides with the «fluoride block of decomposition of carbohydrates», as known in biochemistry. It appeared reasonable to assume that the inhibition of definite links of the cell's chemodynamics, called forth by the paralysants applied, was a condition necessary for delaying the formation of such physiologically active substances of the type of «mediators of chemical excitation», as are acetylcholine and pyruvatecholine. Proceeding from the results of these experiments, as well as from those of the comparative physiological study of the presence of acetylcholine in aneuric organisms, the present author suggested in 1938, independently of the Cambridge biochemists ([5]), a scheme of formation and disintegration of acetylcholine, proceeding in a close relation with the decomposition of carbohydrates, taking place in the cell. This scheme was based on the theoretical assumption that the mechanism of the formation of physiologically active substances which take part in the stimulation of the cell, is intimately connected with the respiratory exchange of the latter. It was further assumed that, as a result of some biocatalytic processes whose nature still remains obscure to us, some individual derivatives of the total metabolic cycle are excluded from it, which are endowed with a high physiological activity and participate directly in the processes of stimulation of the protoplasm, inclusive of nervous excitation, by affecting separate most important links in the chemical processes going on in the protoplasm.

For the first series of our experiments we made use of myographic records, which enabled us to restore the general picture of elimination and weakening of the influence of motor impulses upon the muscular cell, acted upon by certain substances inhibiting the various important links of carbohydrate decomposition. It stands to reason that we could not do otherwise than ascribe the shortening of the amplitude of contractions, which was found, though in a different

measure, to be called forth by the agents inhibiting the chemical transformation of cells in its most important links, to the disturbances in the energetic processes connected with these chemical transformations.

But the sphere of influence of the substances interfering with the normal course of metabolism in nervous and muscular cells (and their intermediate substances) must go far beyond mere disturbances in the energetics of the chemodynamic processes; it must also tell upon such specific processes going on in those structures, as the formation of the so-called neurohumors, and upon the generation of biocurrents, the more so as these two processes require definite sources of energy.

According as we proceeded in our experiments, and also on the ground of the above considerations, we arrived at a point where the study of main electro-physiological indices observed in muscular and nervous formations exposed to metabolic paralysants, has become quite imperative to us.

We investigated the changes that take place in the bioelectric currents of the frog's muscles when acted upon with one of the substances under test, whose effect is to inhibit glycolysis, *viz.* sodium fluoride. The experiments were carried out by myself, together with my co-worker, A. M. Riabinovskaya. As material use was made of neuromuscular preparations of *m. gastrocnemius* and *m. sartorius* of the frog, subjected either to direct, or to indirect stimulation. One of the neuromuscular preparations serving as a control was always kept in Ringer solution, while another identical preparation was immersed into sodium fluoride solution ($M/50$), for a period ranging from 30 to 45 minutes. The muscles were fixed, and the currents of action were led-off from them to a four-cascade amplifier constructed according to the principle of the condenser-rheostat scheme. From the amplifier the currents were led to the tail of a Siemens oscillograph, and recorded upon a moving photopaper-feed-roll. As stimulant crystalline powder of sodium chloride was used, which was applied to the surface of the muscles and the nerve.

The resulting oscillograms were published early in 1941 ([6]). From these oscillograms it may be seen that the effect of sodium fluoride was always to restrict the amplitude of the currents of muscle activity and to reduce the frequency of oscillations, irrespective of whether the stimulation was a direct or an indirect one.

It is of interest to point out in this place that the American writers, Shanes and Brown, in their paper published in February 1942 ([7]), gave an abundant material in support of the existence of changes in the currents of rest in the nerves of the frog, acted upon by paralysants of metabolism, including also sodium fluoride. The changes recorded by these authors were in the same direction as observed in our experiments.

The discovery of a relation between the bioelectric phenomena of rest and excitation, and the biochemical transformations of the protoplasm of the stimulated systems, opens wide prospects for a further inquiry into the nature of this relation.

Numerous data have been accumulated within the recent 10—15 years, pointing, though in a rather indirect manner, to a dependence of the conditions of the generation of bioelectric potential upon the normal course of, say, reducing-oxidizing processes. No direct proof of the generation of bioelectric potential being dependent on chemodynamic conditions could, however, be obtained until quite recently, by applying various inhibitors of the chemodynamics of the cell, acting upon definite links of chemodynamics.

In our earlier oscillographic investigations, published in 1938 by Riabinovskaya and Livanov, from this laboratory, it has been shown that if the skeletal muscle is acted upon by such specific nerve-stimulants as acetylcholine and adrenaline, the oscillogram of the bioelectric currents reveals changes distinctly characteristic of each of these substances ([8]). This is borne out by a detailed analysis of the frequencies and amplitudes of the separate waves of the oscillogram, made by Bernstein and Livanov's method. The periodograms obtained from such

a treatment of the oscillograms make it quite evident that the interference of these substances with the course of the enzymic processes of the cell modifies, in a way differing according to the substance applied, both the intensity and the periodicity of the bioelectric currents. The significant differences in the distribution of waves according to their amplitudes and frequencies, revealed with the aid of the substances tested by us, make us also conclude as to the important rôle played by the biochemical transformations of the cell in the generation of the biocurrents.

However, the question as to the nature of the dependence existing between the so-called «chemical mediators of nervous excitation», as associated with the various links of cellular chemodynamics, on the one hand, and the agents which are responsible for the generation of the bioelectric currents, on the other hand, still remains obscure. We feel inclined to assume that the biocurrents do not always arise in the same way, their generation varying as to the nerve acted upon by the stimulant; we likewise suppose that the different way in which the generation of biocurrents is stimulated according to the varying behaviour of acetylcholine, sympatine and histamine in the general enzyme-chemical reactions of the tissues, is also in full agreement with the recent chemical classification of the various nerves into cholinergical, adrenergical and histaminergical ones.

For the case of cholinergical nerves we must assume that the bioelectric potential arises as a result of a cycle of chemical transformations having in one of its centres acetylcholine with its bonds at the periods of its synthesis, its hydrolysis and its participation in the setting into operation of the cycle of chemical processes characteristic of the excitation of the protoplasm of a given cell (say, in the cycle of transformations of thermo-adenosinphosphoric compounds in the case of the stimulation of a muscle).

According to our idea, the transition from the state of rest of the protoplasm to the state of excitation is characterized by that, during the latter, free acetylcholine is released from the acethylcholinproteid, and, finally, by the hydrolysis of acetylcholine. In our previous note (°) it has already been pointed out that, as may be inferred from our experimental data, the hydrolysis of acetylcholine and, consequently, the liberation of acetic acid, plays an important rôle in the dynamic of the excitation process. Namely, by setting free calcium, magnesium, and potassium from proteinic or lipoid compounds, and causing their transition into an ionized state, acetic acid is creating the conditions responsible for the actual trend of the main energetic process—the decomposition of adenosintriphosphoric acid, either by activizing or inhibiting, through the respective ions of calcium, magnesium or potassium, the enzymic processes associated with the decomposition of adenosinphosphoric compounds.

The physiological rôle of the phase of acetylcholine hydrolysis, which we wish to emphasize here, is not restricted to its connexion, important as it is, with the cycle of transformations of adenosinpolyphosphoric compounds; along with this the hydrolysis can serve as a source of the concentration currents. But it is continuously complicated by free potassium, calcium and magnesium ions being involved in the concentration relations, in the way described above, under participation of acetic acid, or by means of some other metabolic processes still unknown, which take place in a cell acted upon by a nerve stimulant, and also in the cell's molecular structures, containing the said ions in fixed condition.

This metabolic hypothesis advanced by us in order to explain the appearance of the bioelectric potential which is concentrational in its major part, is by no means a novelty: similar ideas have already been expressed by the classics of electrophysiology of the XIX century, *viz.* Sechenow, Hermann. The Russian scientist W. Chagovetz was the first to put this hypothesis upon an experimental basis at the very close of the XIX century; he preceded Loeb and Nernst in giving a physical-chemical interpretation to the process of nervous excitation. In his search for a compound whose participation in the process might account

for the bioelectric phenomena observed in the cell, Chagovetz was forced, for lack of information on the biochemical changes taking place in the cell during excitation, to fix his choice upon the carbonic acid; he found himself obliged, however, to assume the existence of a hypothetic proteino-carbonic acid, in which the carbonic acid was supposed to be in a fixed state, owing to which a constancy of the degree of association is attained.

If we are to operate with the modern interpretations of the nature of the nervous process, we are free to assume that it is the acetic acid released at the loci of excitation in the course of acetylcholine hydrolysis, which is most likely to take part in the arising of the concentration currents, and that it is the concomitant reactions of acetylcholine hydrolysis and synthesis which secure the constancy and continuity of the bioelectric potentials that arise in this way. If viewed at this angle, the existence of concomitant relations between the acetylcholine and the system of adenosinphosphates and the depot of guanidin-phosphoric compounds seems to play a rôle of particular importance in the genesis of the bioelectrical potentials.

It was not until quite recently that Nachmansohn and his co-workers ([10]) have given a brilliant experimental confirmation to the existence of a definite relation between the energy of the electric discharges in the electric organ of the fish, and the energy of decomposition of creatinphosphoric compounds, as connected with the dynamic of the synthesis and decomposition of acetylcholine.

All that has been said above should not be interpreted to mean that the complex nature of the bioelectric potential can be reduced merely to the concentration currents which take their origin in the phase of hydrolysis of acetylcholine. The latter statement is no more than an illustration of the part played by the enzymo-chemical processes in the arising of the potentials of concentration origin. But more complicated conditions of generation of bioelectrical potentials, involving the formation and disappearance of polarized microstructures of the cell, may be underlain by definite enzymochemical processes. This is the only possible way to account for the variability of both the high-voltage and low-voltage component of the bioelectric currents, observed in the case of disturbances in the normal course of cell metabolism.

We believe that the efforts of both physiologists and biochemists should be concentrated at the present time upon the problems of the relations existing between the different links of enzymochemica transformations and the decisive factors of the generation of biocurrents (polarization phenomena, in particular); the solution of these problems attained in the way traced by experimental research is of great theoretical interest and practical importance.

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PHYSIOLOGY

CARBONIC ANHYDRASE OF BLOOD AS AFFECTED BY REDUCED ATMOSPHERIC PRESSURE *

By E. G. CHENYKAEVA

(Communicated by L. A. Orbeli, Member of the Academy, 3. VIII. 1944)

Carbonic anhydrase, when studied in animals or men under conditions of reduced atmospheric pressure, is of interest both from practical and theoretical point of view. In its practical aspect it is connected with problems of mountain and altitude sicknesses; whereas in point of theory it is associated with the problem of the so-called «tissue adaptation» of the body to anoxia conditions. Paul Bert was the first to call attention to the possibility of the tissue agent being operative in the adaptation to oxygen deficiency; since his time nearly all authors engaged in this subject have spoken of «tissue adaptation», but no positive proof or convincing evidence has been given so far. The study of the changes that take place in tissue and blood enzymes under anoxic conditions may prove useful for a better understanding of «tissue adaptation».

Kreps and co-workers (1) have shown that various disturbances in respiration conditions may bring about certain changes in the activity of carbonic anhydrase.

The present paper deals with such changes in the carbonic anhydrase of blood as are observed in men and animals when these are subjected to reduced barometric pressure in a low-pressure chamber.

Before we proceed to the experimental data, two points are to be mentioned.

1. The reduction of atmospheric pressure is an agent of considerable complexity. Of prime importance is the reduced partial pressure of oxygen, and the resulting anoxia. The anoxia, causing overventilation of the lungs, leads to a loss of carbon dioxide, to a state of acapnia. Acapnia itself produces certain shifts in the acid-base balance of the body. It is quite possible that anoxia, acapnia and the disturbances in the buffer systems of the blood may individually produce some special influence of their own upon the carbonic anhydrase of blood and tissues.

2. Observations on men and animals in low-pressure chambers show that even short exposures to reduced atmospheric pressure bring about some adaptation phenomena.

Several phases may be observed to succeed one another in the behaviour of animals or men, as well as in the sensations of the subjects under experiment. Changes in carbonic anhydrase are possibly phasic, too.

It is clear, therefore, that no simple relations are to be expected between the reduction of atmospheric pressure and carbonic anhydrase of the body.

The results of our experiments are shown in Tables 1, 2 and 3. They have been obtained with pressures corresponding to altitudes from 4500 up to 9000 and exposures from 15 min. up to 4 hours.

* The work was carried on at the laboratory of the aviation medicine of the Kirov Military Medical Academy. Thanks are due to the head of laboratory Prof. M. L. Brestkin for the interest and the part he took in this work.

Carbonic anhydrase was measured in blood obtained in men by a finger prick, and in dogs, by a puncture of a small artery or vein of the ear. The determination of activity was made by the method of Brinkman, according to a technique developed by Kreps and Chenykaeva (1).

The data presented show that all experiments may be divided into two groups: 1) ascent to an altitude of 4000—5000 m and 2) ascent to altitudes from 8000 to 9000 m.

All experiments with men, and a part of experiments on dogs make up the first group.

The degree of anoxia was a slight one, the behaviour of men and dogs under experiment was quite normal, and no abnormal sensations, or only very slight ones, were experienced by the subjects.

In the experiments of the second group the anoxia was severe; the behaviour of dogs was abnormal, the animals being under extreme conditions.

Table 1

Altitude up to 5000 metres

No.	Altitude, in m	Exposure time, in hours	Subject	Carbonic anhydrase			Erythrocytes in thousands	
				beginning	after 1 h.30′	end	beginning	end
1	4500	3	E. M. K.	3.7	3.7	5.5	—	—
2	5000	3	A. A. S.	3.4	3.7	4.0	4510	4380
3	5000	3	N. A. V.	2.0	2.8	3.0	4160	4340
4	5000	4	Dog Kashtan	5.8	—	6.2	—	—
5	5000	4	» Norka	4.8	—	6.6	—	—
6	4500	3	» Kashtan	5.8	6.3	6.4	—	—
7	4500	3	» Norka	6.0	6.0	7.0	—	—

In all the experiments of the first group (in men and dogs) carbonic anhydrase was markedly increased at the end of the exposure as compared to the initial value.

The increase in carbonic anhydrase was far beyond the possible error of determination.

The rate of the increase observed in carbonic anhydrase showed individual variation. In some subjects and animals (N. A. V., dog Kashtan) the shift reached its maximum within the first 1.5 hours of exposure, whereas in others (E. M. K., dog Norka) the maximum change developed within the later part of the experiment; some of the subjects showed a gradual increase in carbonic anhydrase with time (A. A. S.).

Considerable individual variation should also be detected in the rate of the return to normal values, after the subjects or animals were removed from the low-pressure chamber.

Red-blood cell counts were made in two experiments of this group. In one experiment a slight increase was observed, in another, a slight decrease.

The changes in carbonic anhydrase activity could not be explained by the changes in red-blood cells, since the rise in carbonic anhydrase was of a much greater degree of magnitude than the changes in erythrocytes. We presume that the mechanism of the changes in carbonic anhydrase, when exposed to reduced barometric pressure, is to be sought along the lines of activation and inactivation of the enzyme.

In the second series of experiments (severe anoxia, altitudes 8000—9000 m, corresponding to oxygen percentages of 6.5—7.5) distinction must be made between experiments of short exposure (15—30 min.) and those of long duration (1—2 hours).

Table 2

Altitude, 9000 m. Exposure, 15—30 minutes

No.	Exposure time, in min.	Subject, dog	Carbonic anhydrase		Erythrocytes, in thousands	
			beginning	end	beginning	end
8	15	Norka	5.0	4.4	—	—
9	»	Bobik	5.2	4.2	—	—
10	»	Sharik	4.3	3.5	—	—
11	»	Bobik	4.1	3.5	—	—
12	»	Norka	4.0	4.5	—	—
13	30	Kashtan	6.0	5.2	5110	3320
14	»	Norka	6.9	5.2	4780	4300
15	»	Bobik	5.2	4.3	—	5180

Severe anoxia of short duration (Table 2) was followed, as a rule, by a decrease in carbonic anhydrase. In experiments of this series the animals were usually lying on their side, panting heavily, and when trying to rise or to change their place, they were unable to make more than two or three steps, tottering on their feet.

In 8 experiments of this series carbonic anhydrase showed a marked decrease, and only in one experiment (No. 12) there was a slight increase. It may be mentioned that in this single experiment the initial figure of carbonic anhydrase was unusually low, and even after the observed rise it did not reach the niveau usual for the dog in question.

In two experiments (Nos. 13 and 14) the count of the red cells showed a decrease, but it is difficult to ascertain whether the fall in carbonic anhydrase should be accounted for by this decrease in erythrocytes. A sudden rise in the red cells of blood may be understood as due to the contraction of the spleen. But it is difficult to give a satisfactory explanation to a sudden fall in erythrocytes in the circulating blood.

Severe anoxia of long duration (Table 3) showed no uniform results. In three cases the anoxia conditions led to a rise in carbonic anhydrase, in three other cases, to a fall, and in one case carbonic anhydrase remained practically unaltered.

Table 3

Altitude, 8000—9000 m. Exposure 1—2 hours

No.	Altitude, in m	Exposure, in hours	Subject, dog	Carbonic anhydrase		Erythrocytes, in thousands	
				beginning	end	beginning	end
16	9000	1	Norka	6.2	7.2	4930	5330
17	»	»	Ryzhik	6.7	7.2	—	—
18	»	»	Sharik	6.0	7.3	5370	4840
19	8000	2	Bobik	3.3	3.5	4370	—
20	»	»	Kashtan	7.6	7.1	—	5580
21	»	1½	Norka	6.4	5.6	—	—
22	»	»	Kashtan	7.0	6.5	—	—

The behaviour of the animals was not uniform either. In some cases the animals remained till the end of the experiment in a bad state, not differing from their state at the beginning of severe anoxia. In other cases the dogs showed outer

signs of a developing adaptation as the time went on. They assumed the usual posture of a sleeping dog, were able to walk about the chamber in a normal way.

In other words, they were already in another phase, as compared to the beginning of the experiment.

The count of the red cells did not give concordant results in this series of experiments.

In some cases there was a rise, in others, a decrease, sometimes following the change in carbonic anhydrase, sometimes going in the opposite direction.

The data obtained may be summarized as follows:

1. A slight degree of anoxia, which does not provoke any intense hyperventilation of the lungs with a corresponding acapnia, is in all cases followed by a rise in carbonic anhydrase.

2. A more severe anoxia with intense hyperventilation brings about a decrease in carbonic anhydrase.

3. When the action of reduced barometric pressure of such intensity is continued, the decrease in carbonic anhydrase may give place to an increase. It is probable that this change should be considered as a manifestation of various phases in the organism's reaction.

We suppose the observed changes in carbonic anhydrase be of an adaptive nature.

The organism's reaction may be of a various nature, according to whether anoxia or acapnia is the agent prevailing at a given moment (anoxia or acapnia).

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